ISBN: 978-92-76-09684-9
ISSN: 1831-9424
JRC N°: 65614
EUR 24882 EN

ESARDA
European Safeguards Research and Development Association

Hungary
BudaPest
ESARDA 2011

33rd Annual Meeting

Symposium on safeguards and nuclear material management

May 16–20, 2011 | Budapest, Hungary

JRC
EUROPEAN COMMISSION

Publications Office of the European Union,
Luxembourg ISBN 978-92-76-09684-9
doi: 10.2760/07768

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01 Plenary Session

Application of nuclear energy in Hungary

Dr. József Rónaky

Director General

Hungarian Atomic Energy Authority

The Hungarian nuclear profile

- Fuel cycle
  - Uranium mining (ore) – nowadays (extraction from water)
  - Uranium ore selection – nowadays (yellow cake)
  - Ore exported
    - No conversion
    - No enrichment
  - Fuel assemblies imported
  - Power reactors, research reactor, training reactor
  - Spent fuel exported
  - Interim storage of spent fuel (Spent fuel ponds, SFISF)
    - No reprocessing
    - Research for final depository

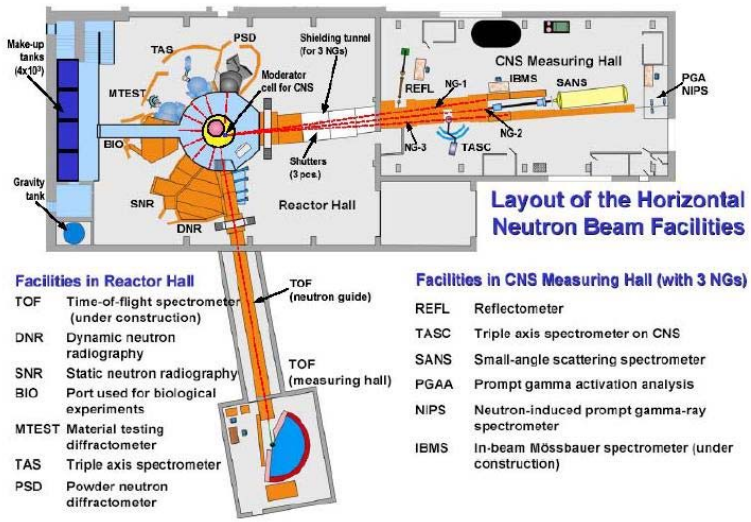
Nuclear facilities and other users 1/6

- **Uranium mine**
  - Study of uranium density in the country (1949-1955)
  - Mecsek mountains, close to south border
  - Construction (1955-1957)
  - First ore export to SU (1958)
  - First yellow cake export - after chemical treatment (1962)
  - Closing of the mine (1997)
    - ~ 40 years – 23 000 t U
  - Uranium extraction from ground water
    - ~ 2-3 t U / year

http://www.mecsekoko.hu

Nuclear facilities and other users 2/6

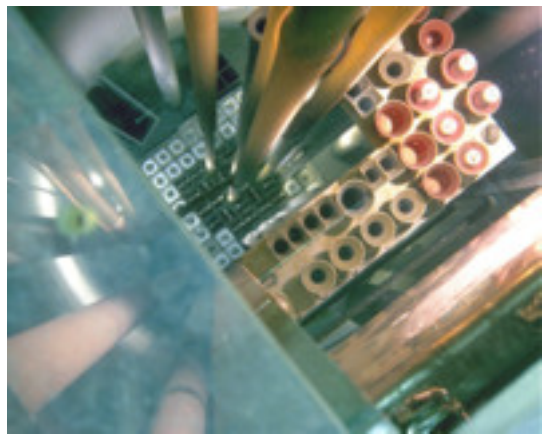
- **KFKI Budapest Research Reactor**
  - First criticality (1959) – 2 MWth
  - Power upgrade (1967) – 5 MWth
  - Total reconstruction (1986-1993) – 10 MWth
  - Purpose:
    - Training, neutron-physics, reactor-physics research, radioactive isotope production
  - Reactor
    - Tank type (water-water, beryllium reflector)
    - Uranium fuel
    - Enrichment: LEU, HEU
  - Spent fuel pool
  - Fresh fuel
  - Zero power critical systems (1959-1990)



www.kfki.hu

Nuclear facilities and other users 3/6

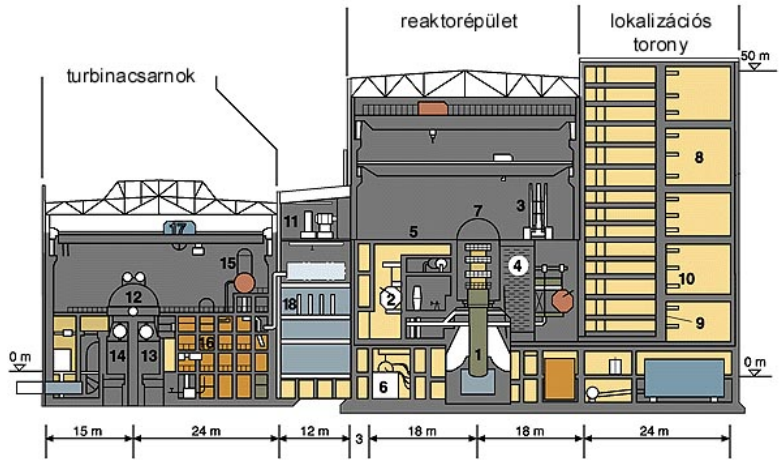
- **TUB Training Reactor**
  - Construction (1969-1971)
  - First criticality (1971) – 10 kWth
  - Power upgrade – 100 kWth
  - Purpose:
    - Training, neutron-physics, reactor-physics research, instrumentation development, activation analytics, radiochemistry
  - Reactor
    - Pool type (water-water, graphite reflector)
    - Uranium fuel
    - Enrichment 10%
    - 23-24 assemblies (in Al cladding)
  - Low burn-up
  - Fresh fuel



www.reak.bme.hu

Nuclear facilities and other users 4/6

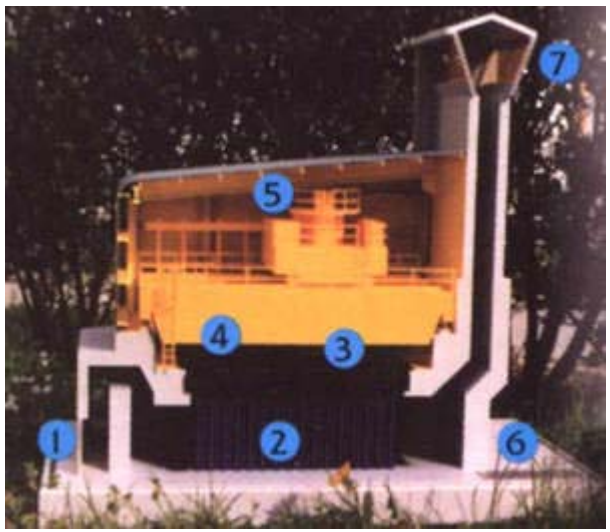
- **Paks Nuclear Power Plant**
  - 110 km (southward) from Budapest
  - 4 units (VVER-440/213)
  - Construction (1975-1987)
  - Power upgrade – 500 MWe
  - Reactor
    - PWR (water-water)
    - Uranium fuel
    - Enrichment 3,8%
    - 349 assemblies
  - Spent fuel pools
  - Fresh fuel storage
  - 2009: Parliament ‘s licence in principle to commence prepration activities for new units



www.atomeromu.hu

Nuclear facilities and other users 5/6

- **Spent Fuel Interim Storage Facility**
  - 110 km (southward) from Budapest
  - Construction (1995-)
  - Modular vault type
  - 50 years interim storage
  - Passive air cooling



Nuclear facilities and other users 6/6

- **Universities**
  - Nuclear sources for research
  - Debrecen
  - Budapest (ELTE, TUB)
  - Veszprém
- **Research Institutes**
  - Nuclear sources for research
  - KFKI AEKI (Budapest)
  - ATOMKI (Debrecen)
  - VEIKI (Budapest)
  - IKI *www.iki.kfki.hu*
  - *OSSKI*
- **Other companies**
  - Industrial applications

7 MBA (among them HU-C for small users)
11 AP sites

Nuclear materials 1/2

- Natural uranium
  - In almost all facilities (chemicals, etalons)
  - Mecsek ÖKO Co. (extraction from groundwater)
  - Underground (20 000 tons)
- Enriched uranium
  - Fresh and irradiated reactor fuels
  - Enriched research materials (small amount, calibration sources, research samples)
- U-233
  - Few mg-s in research labs
- Depleted uranium
  - Only import (by-product of enrichment)
  - Container walls, radiation protection, counter-weights, ballasts

Nuclear materials 2/2

- Plutonium-239
  - Spent fuel (7-8 tons)
  - Pu-BE sources – (measurement of real content in IKI) (5-6 kg)
  - In smoke detectors (early application)
  - Etalons, metal samples
- Thorium
  - Gas-lighting – gas-skirt
  - GE metal-halogen lamps cathode-surface cover

Nuclear research

- Application of nuclear and radioactive materials
  - Industrial
  - Medical
  - Research
- No institutional research in the fields of
  - Fuel preparation
  - Fuel design
  - Fuel reprocessing
- Major areas in nuclear research
  - Reactor safety analyses and research
  - Training demonstrations
  - Instrument calibration
  - Particle accelerators

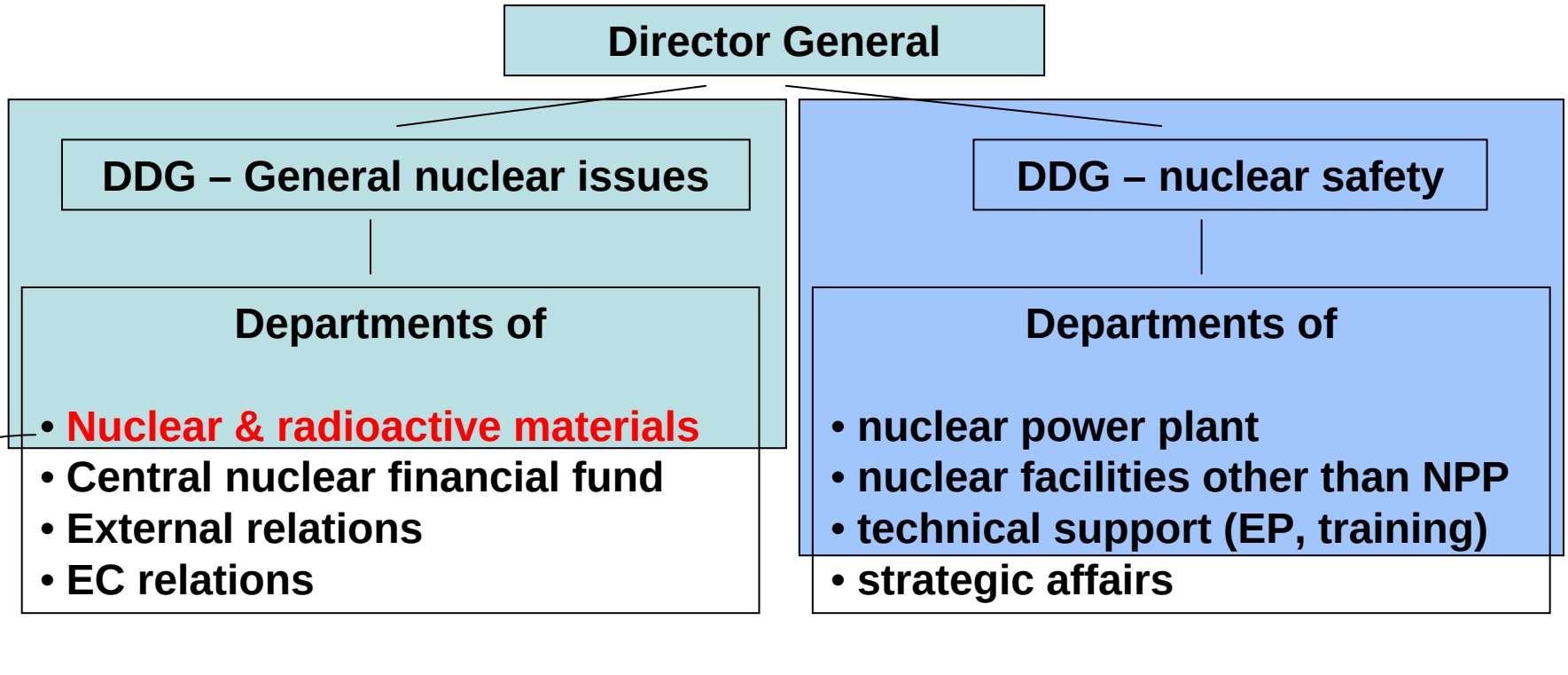
Inspection regimes

- Non-proliferation treaty
- Comprehensive Safeguards Agreement
- Additional Protocol
- EURATOM-IAEA-Hungary 3-lateral agreement
- Comprehensive test ban treaty
- Nuclear Suppliers Group
- Zangger Committee
- Convention on Physical protection

- National legislation

Regulatory scheme

Hungarian Atomic Energy Authority



nuclear safeguards, nuclear export-import control, physical protection of NM, CTBTO central registry of RM (sources & waste), transport, packaging, illicit trafficking

Regulatory supervision

- Reporting, data collection and processing
- Licensing (new element from 2007)
- Inspections

Reporting, data collection and processing

- Central register of NM
- Local registers of NM
- Daily, monthly, half-yearly, yearly reports to regulator
- Eventual reports to regulator

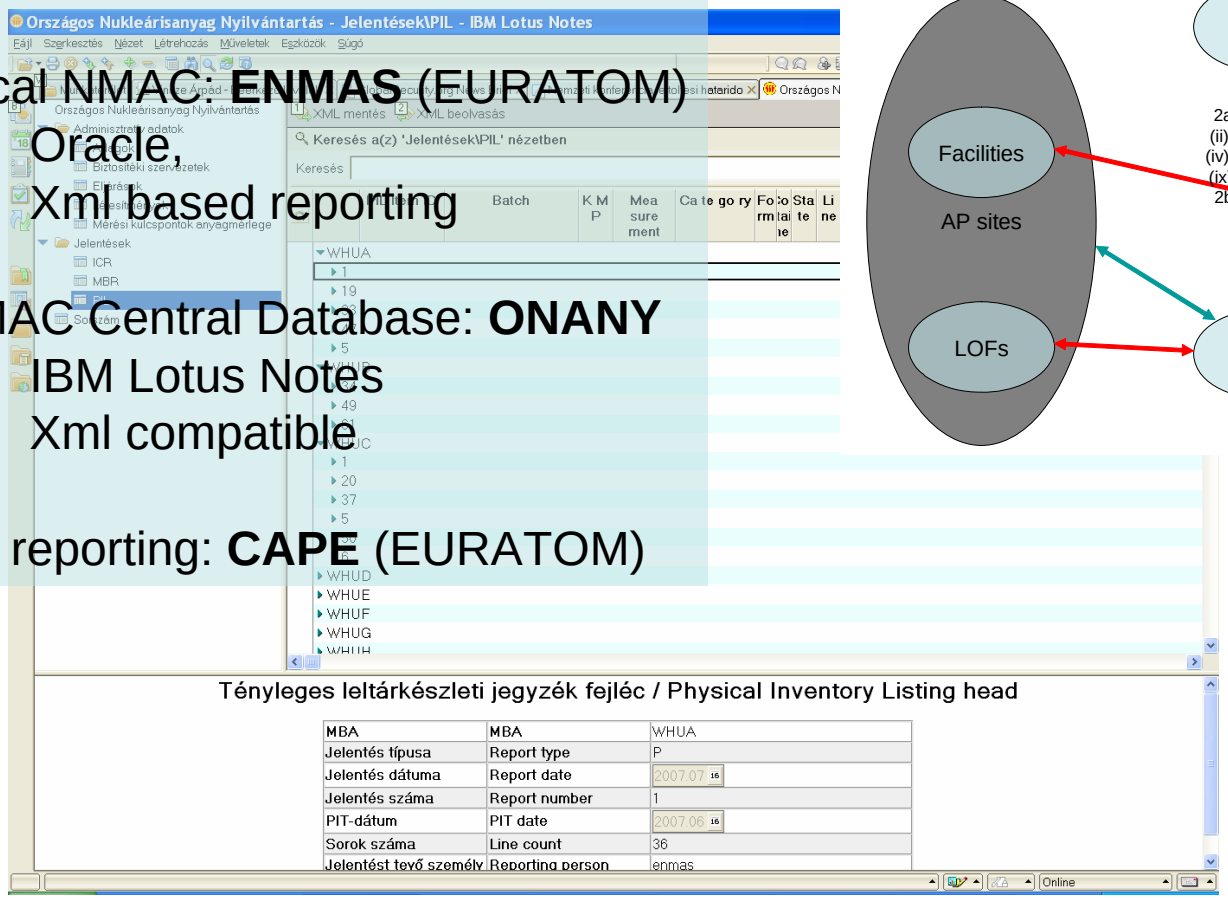
- Reports to EURATOM and IAEA

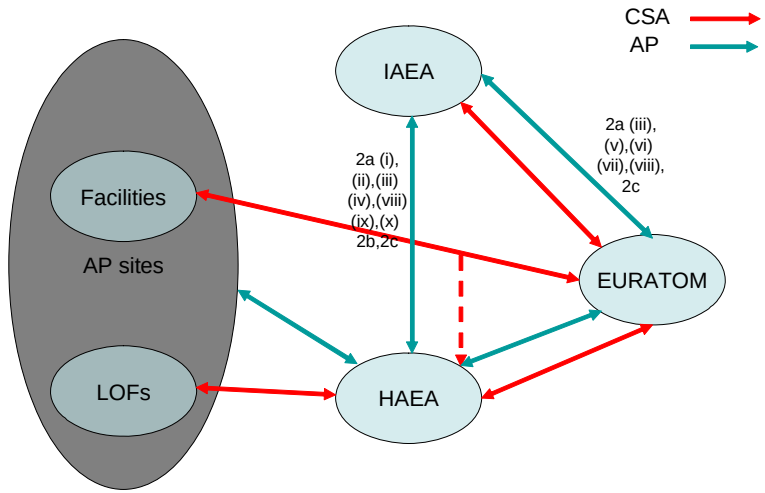
in accordance with EURATOM
and IAEA requirements

Data Processing Scheme

SSAC Information System:

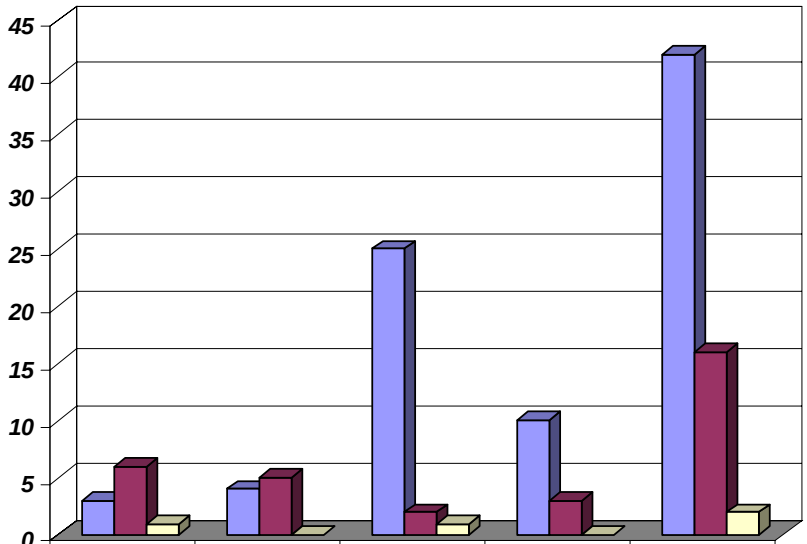
1. Local NMAC: **ENMAS** (EURATOM)
   - Oracle,
   - Xml based reporting
2. NMAC Central Database: **ONANY**
   - IBM Lotus Notes
   - Xml compatible
3. AP reporting: **CAPE** (EURATOM)





Licensing scheme

- For possessing nuclear material, for starting nuclear activity (first license)
- For modifications (modification license)
- For export-import of nuclear and dual use materials and equipment (transport license)
- For termination of safeguards (exemption license)



	2007	2008	2009	2010	Sum
First	3	4	25	10	42
Mod	6	5	2	3	16
Trans	1	0	1	0	2

•Requirements are harmonized with large and small users (graded approach)
•30 days deadline for regulator (may be extended with 30 days once more)
•Regulatory resolution (licensing and enforcement)

Verification Scheme

Necessity to draw independent conclusion

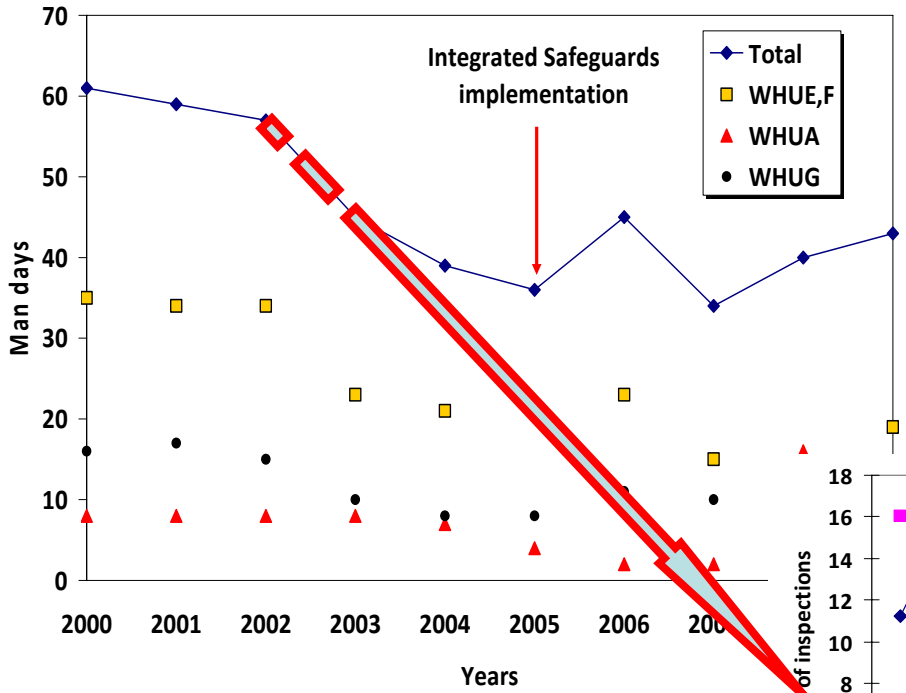
- Planned in advance based on facilities' sg relevant activity schedule
- mostly carried out simultaneously with the inspection of the IAEA/Euratom,
- relies on the containment and surveillance systems installed at the facilities by the IAEA/Euratom,
- relies on the measurement carried out by the IAEA/Euratom with its equipment

BUT

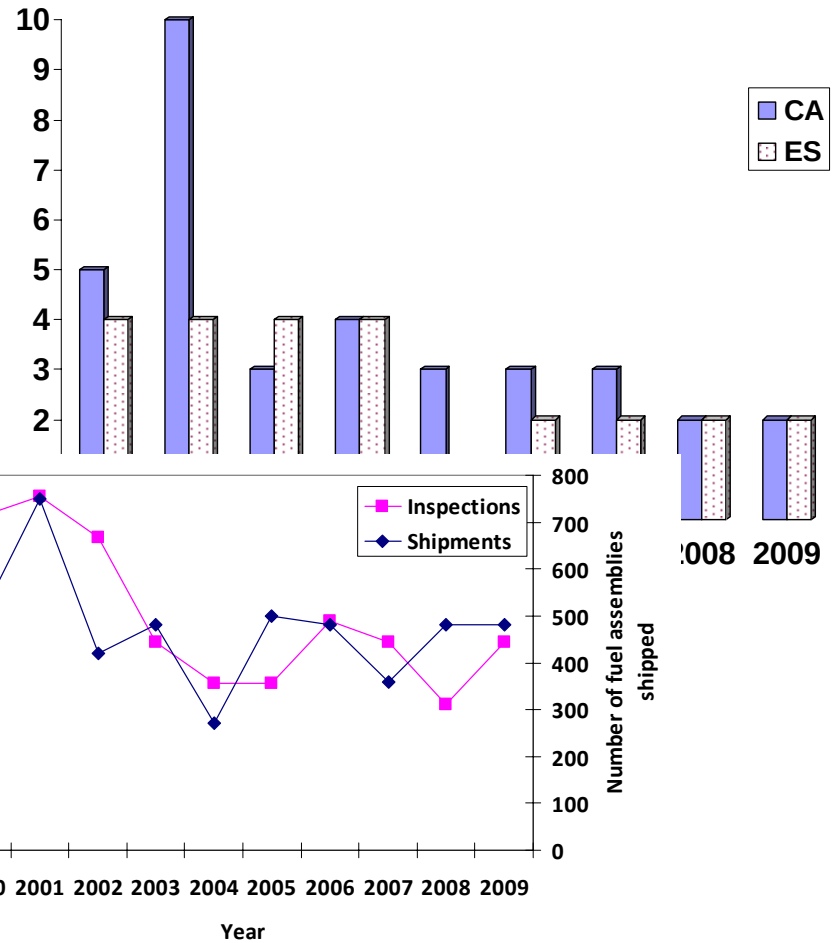
- Independent inspections and measurements

Site inspections - international

Traditional safeguards



Complementary Access



National Implementation Philosophy

- risk based approach
- harmonization with safety
  (e.g.: core verifications, PuBe sources)
- independent measurements
- all inspection results are stored in a database and analysed
- support for SG R&D
  (8-10 projects per year)

Regulatory supervision - conclusions

- Safeguards regulatory body
  - Same importance as nuclear safety (both for regulator and operators)
  - Safeguards aspects are considered in details
  - Strict deadlines according to national laws
  - Enforcement capability
  - Independent conclusion to Hungarian government

Thank you for your attention!

The Future of Safeguards: Adapting to change

Herman Nackaerts

Deputy Director General and Head of Department of Safeguards,

IAEA

Since its creation in 1957, the Agency has been tasked with verifying that States are in compliance with their commitments to use nuclear material, equipment and technology for peaceful, non-explosive purposes.

The yardstick by which we judge the effectiveness of our safeguards system is very simple: Are we able to detect early the diversion of nuclear material from peaceful nuclear activities – and thereby to deter any State that might be contemplating such an act? And are we able to provide credible assurances that States do not possess undeclared nuclear material or are not conducting undeclared nuclear activities – and thereby to allay the fears of other States about potential proliferation?

In attempting to answer those questions, my basic thesis is that the present safeguards system is not as effective as it could be, and that we are not applying our resources in the most efficient manner. Of course, there are reasons for the way in which we implement safeguards today: the concept and approaches underlying safeguards – largely the result of political and technical compromises – were valid when they were established. But are they still valid now?

Today I want to set out why – if we are to continue to fulfil our mandate effectively – the Agency needs to modernise its safeguards system and needs to implement safeguards in a more focussed way.

1. So, what is wrong with the present system?

After the weakness of the traditional safeguards system was glaringly exposed in the early 1990s with the discovery of Iraq's clandestine nuclear weapons programme, it was recognised that a system almost exclusively focused on the verification of declared nuclear material was totally inadequate.

The message was clearly understood – the Agency began a programme for strengthened safeguards which involved the addition of new tools and legal authorities to our verification toolbox, including the introduction of the additional protocol. This effort was directed at improving the effectiveness of the safeguards' system. In addition, it became clear that the implementation and evaulation of safeguards would have to look at the State as a whole, and not just at the sum of its individual facilities. Hence, the Agency developed the 'State-level concept' and introduced State-level integrated safeguards approaches in those countries that had an additional protocol and had obtained the broader conclusion. The main focus of this effort was to improve efficiency without undermining effectiveness.

These initiatives are commendable: however, their inherent strengths have not been fully exploited. For example, while the State-level concept is being applied to the evaluation process it has not resulted in significant change to safeguards implementation in the field. The determination of this effort continues to be based very much upon the traditional 'safeguards criteria' which, in turn, is focused on the nature and quantity of nuclear material and the types of nuclear facilities within a State. Even under State-level integrated safeguards approaches, the level of implementation was largely determined by criteria-relevant considerations, such as adjustment to timeliness and quantity goals. The system remained largely prescriptive, driven by pre-determined criteria and almost exclusively focused at the facility level. "Satisfying the criteria" was in danger of becoming an end in itself rather than addressing the underlying

objective of the work – that is, whether there were any genuine proliferation concerns.

The Libya case was a stark demonstration that nuclear technology had become available on the black market: that there were covert nuclear supply networks that could supply the most sensitive nuclear technology – including designs for a weapon itself.

What the Libya case also highlighted was the emergence of a new 'non-State' threat, about which the Agency needed to become much more aware.

Then there was the case of the undeclared laser enrichment experiments in the Republic of Korea, which also reflected important gaps in the overall system. Here, the Republic of Korea's own State System for Accountancy and Control (SSAC) was unaware that such activities had been taking place at the national nuclear research institute.

108 of our Member States have now put in place an additional protocol. We welcome this development. The additional protocol provides us with the ways and means to verify the completeness of a State's declarations through access to a wider range of information and to more locations within the State. This is a positive development. However, of those States yet to conclude an additional protocol, seven[1] have significant nuclear activities. In particular, the absence of an additional protocol in Iran and Syria, is severely restricting our ability to look beyond declared facilities and activities. In all States without an additional protocol the Agency is not in a position to fully implement its mandate and fulfil its obligation to confirm that there are no undeclared nuclear material and activities in these countries.

[1] Algeria, Argentina, Brazil, Egypt, Iran, Syria, and Venezuela.

Even without the additional protocol, a comprehensive safeguards agreement contains provisions relevant to the detection of possible undeclared activities. The problem is that, over the years, some of these provisions have been interpreted in a rather restrictive manner, and neglected. An obvious example is the special inspection, which the Agency can conduct, inter alia, if information gained from routine inspections and explanations from the State are inadequate.

2. How we need to change

If we want the safeguards system to become more effective, to better address our basic obligations of deterrence and early detection and to respond to the changing landscape, we need to significantly change the way we undertake our work. It is important to note that the changes that I will be referring to do not require any change to our existing legal authoritity.

Clearly, the quantity of nuclear material in a State, or even the size of its fuel cycle, constitute poor indicators of the proliferation risk posed by that country. They should not, therefore, be the only parameters that determine the level of effort the Agency expends on a particular country. Indeed, as I mentioned earlier, previous cases of violations or clandestine nuclear weapons development occurred in countries with a limited nuclear fuel cycle, and involved previously exempted or undeclared nuclear material.

It is also clear that the best time to act against the diversion of nuclear energy to non-peaceful purposes is before the activity commences: deterrence is much more effective than detection.

Therefore, the time has come to fully and properly implement a safeguards system that truly considers the State as a whole; that is less predictable and therefore has a higher deterrence value; that is more focussed on areas of

concern; and that is flexible so it can easily adapt to a changing situation. When determining the level of verification activities in the field we should consider other factors that go beyond traditional considerations of the type and quantity of nuclear material.

Looking at the State as a whole, we need to be in position to clearly understand its overall nuclear profile in order to confirm the peaceful nature of its nuclear programme. In using all of the information available to us, we want to be able to develop a customised approach – one that takes into account all of the State-specific factors. The Agency's safeguards system needs to be flexible, driven by outcomes, supported by transparent and consistently applied processes, and judged against our overall aim to deter proliferation.

This has attractions for both the State and the Agency. The Agency can focus its verification where it really matters, while still retaining the option of shifting that focus or conducting verification activities elsewhere if circumstances demand it. The State, on the other hand, when it is cooperating well and is providing correct and timely information isn't overburdened by unnecessary inspection activities. Precious resources can be conserved without the quality of verification being compromised. On the contrary, the overall effectiveness of the system will be enhanced.

3. Challenges

But changing the approach to implementing the safeguards system in this direction will involve a number of challenges that need to be addressed.

First, we need to be very clear to our Member States that we are not proposing to implement different safeguards; rather, we want to implement safeguards differently. Nor do we intend to discriminate against certain States or categories

of State. All States will remain subject to the same rules and overall objectives as before and to the agreements they have signed with the Agency.

If the Agency's implementation of safeguards for any given State is uniquely customised – driven by all available safeguards-relevant information, we will need to be clear and transparent about how our analysis of this information led us to take certain safeguards actions in that State as a result. The Agency will need to be able to justify those actions: a process that will be more complex than when we were acting in accordance with fixed, pre-determined criteria.

We will need to be able to persuade Member States that what we are proposing is not only non-discriminatory, but also that it is the right thing to do – that it will improve, not undermine, the quality of our safeguards findings and conclusions. We will have to demonstrate that while the new system will focus more on those trying to break the rules or avoid their responsibilities, the overall inspection burden on everyone else will be reduced.

The skill sets within the Department of Safeguards and of the inspectors themselves need to be broadened. Instead of just being accountants, our safeguards practitioners also need to be investigators and analysts.

We will also use this opportunity to look again at our legal basis, to review whether we cannot make better use of the tools at our disposal – contained within the safeguards agreement – to have better access to information, locations and people, specifically in countries that are not implementing an additional protocol.

The ultimate challenge will be to demonstrate that these changes will bring about a system that delivers strong conclusions and provides credible non-proliferation assurances to the world.

4. Implementing Change

In some ways, what I am proposing is not new. It is a natural continuation of a process that began in the early 1990s when strengthening measures were agreed through the Programme 93+2 and the subsequent introduction of the additional protocol.

For the growing number of States under integrated safeguards, the State-level approach has already been developed and applied. It includes, in theory at least, all the elements described previously. However, while the theory is in place, in practice the concept has not yet been fully implemented, and our activities remain largely criteria driven and prescriptive. State-level evaluation activities are not fully integrated with the processes to determine verification effort. It is now time to put that right.

Because verification of a State's declaration is essential, nuclear material accountancy will remain an integral part of the safeguards system and the backbone of our activities.

Nevertheless, we do need to look at whether the level of verification we currently have in place is really needed in all cases. We believe that the systematic use of the State evaluation process and its resulting State-level approach, in which all relevant State-specific factors are taken into account, can better qualify the non-proliferation credentials of a State and accordingly generate verification levels that are more commensurate with the risks.

The State-level approach should be applied to all States with a safeguards agreement in force and be based on a comprehensive evaluation of all available safeguards-relevant information. Through a collaborative process, this information then needs to be analysed in order to build a comprehensive picture of a State's nuclear activities. It is upon an evaluation of all this information –

the State evaluation process – that we plan and implement our verification activities and ultimately draw our safeguards conclusions for each State.

This State evaluation process is a dynamic, robust and iterative process, the outcome of which is used as the basis for planning subsequent safeguards activities. The results of those activities, in turn, are themselves assessed and any follow-up actions identified – for example, whether additional information is required or further verification activities need to be conducted. So, if the information and evaluation change, so does our safeguards' approach in that particular State. Thus, safeguards implementation at the State level is fully "information driven".

Director-General Amano has fully endorsed our change programme which is already underway. We have set ourselves an ambitious schedule – we want all of these changes to have been implemented by the end of 2012. For this to happen we will need the support of all our stakeholders.

5. What does it mean to stakeholders?

Nuclear non-proliferation is a collective global effort, which is more likely to succeed when all members of the international community work together. Likewise, the IAEA is more likely to be successful when it works in partnership with States. Hence, we will reach out to States to increase the voluntary sharing of safeguards-relevant information: this will particularly help to strengthen our ability to detect undeclared activities.

We also rely on States' support in other areas, particularly in the day-to-day implementation of safeguards. We will work to help ensure that States have competent safeguards authorities and we will support States in enhancing the effectiveness of their State systems of accounting for and control of nuclear

material (SSACs). Where possible, we will then make greater use of them – thereby also gaining greater efficiencies in the use of our resources. This is of particular interest to us here in Europe, where the Agency has enjoyed lengthy and active cooperation with the regional Euratom safeguards system. The change that we are proposing is an opportunity to review the partnership we have with Euratom and to investigate how we can establish synergies and enhance the cooperation between the two organisations.

The safeguards system needs to be transparent and understandable – internally to those who are implementing it, as well as externally to Member States and the general public. Therefore, we will seek to improve the openness and quality of the Agency's reporting on safeguards and verification matters; to build States' knowledge of the processes for drawing safeguards conclusions and thereby enhance their confidence in the Agency's assurances.

Conclusion

So, the challenges are numerous and not insignificant. But by being responsive to change and flexible in application, and by focusing on areas of real proliferation concern, we can ensure the more efficient implementation of more effective safeguards. Only by adapting the safeguards system and by applying safeguards more intelligently, will we be able to continue to provide the international community with credible assurances that States are in compliance with their safeguards commitments.

I invite you to join us as we embark – not on a revolution – but on a process of 'accelerated evolution' towards smarter safeguards.

Thank you.

33rd ESARDA Annual Meeting

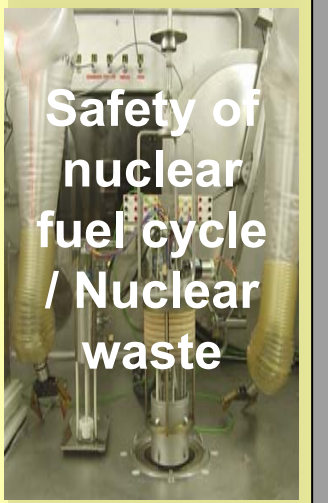
Budapest, May 16-20, 2011

JRC's Grand Challenges in Nuclear Safeguards and Security

Th. Fanghänel
Institute for Transuranium Elements
Joint Research Centre, European Commission

Structure of the nuclear programme

Reference Centre for
policy makers, stakeholders and citizens
in the nuclear field



Underpinning Science: atomistic and molecular process understanding of fundamental phenomena

Nuclear Safeguards and Security @ JRC

– **Activities at 2 institutes of the JRC**

IRMM, Geel (*Institute for Reference Materials and Measurements)*

ITU, Karlsruhe & Ispra (*Institute for Transuranium Elements)*

Examples of JRC's activities in nuclear safeguards & security typical of present and future challenges, addressing:

– **Support Safeguards authorities Euratom (DG ENER) and IAEA**

R&D; S/T operational support; Training (and education)

– **Advanced nuclear fuel cycles: Safeguards & Proliferation Resistance**

– **Non-proliferation & proliferation concerns**: absence of undeclared activities, particle analysis, trade analysis

Nuclear Safeguards and Security @ JRC

– **Prevention, detection and response to illicit acts involving nuclear materials**

Equipment development, testing and validation

Nuclear forensics scientific advancements

– **Training and Education**

➢ EU nuclear SECurity TRAining Centre; European School for Nuclear Safety and Security

– **International collaborations →**

International Collaboration / Major Nuclear Safeguards & Security Actors in connection with JRC

IAEA.org International Atomic Energy Agency

UN agency, 1957
Safeguards and Verification
Non proliferation, Additional protocol
Nuclear Security / IEC

EURATOM
Rome treaty 1957
Chapter VII on nuclear safeguards

DG-ENERGY
Nuclear Energy and Safeguards

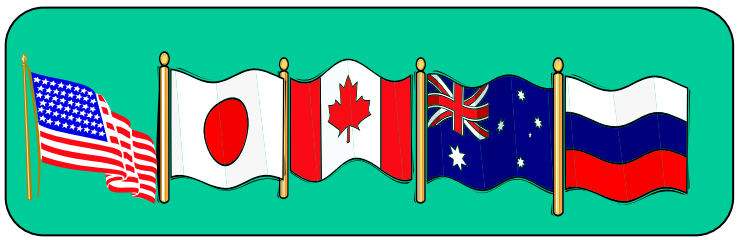
EU Member States

DG DEVCO
Instrument for Stability and Nuclear Safety

JRC
(Joint Research Centre)
Nuclear Security
R&D, T&S support

DG HOME
EU CBRN action plan

EEAS



Other DG's
TRADE
TAXUD
ENTR
....

Trad'l Safeguards: On-Site Laboratories

JRC performs nuclear analytical measurements on behalf of Euratom Safeguards, Luxembourg, independently of the operators.

Sellafield Ltd

On-Site Laboratory (OSL)

Institute for Transuranium Elements

AREVA NC

At La Hague
Laboratoire Sur-Site (LSS)

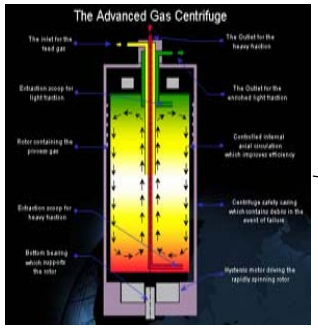
15 June 2010

The target uncertainties for the measurement of U/Pu in the input/output of the reprocessing plants are about 0.1% to 0.6%

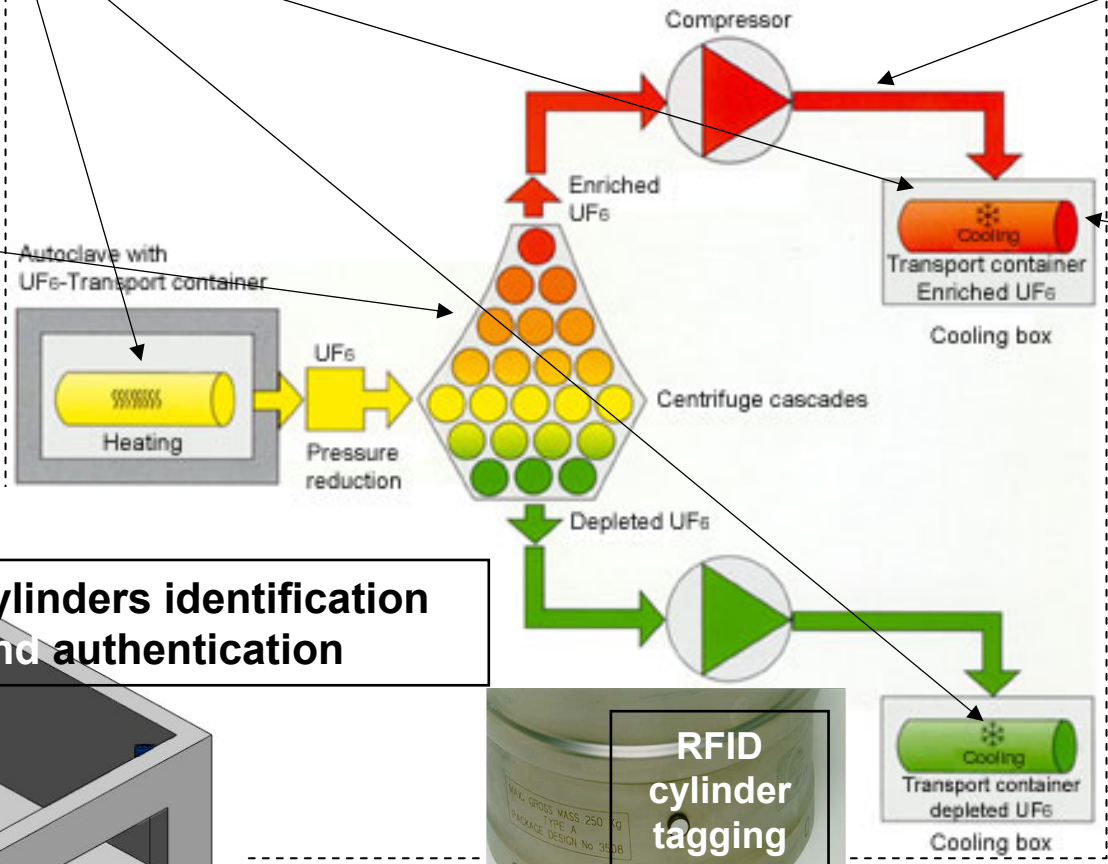
ESARDA / IAEA target values met under daily operation conditions

Integrated approach to safeguards of GCEP

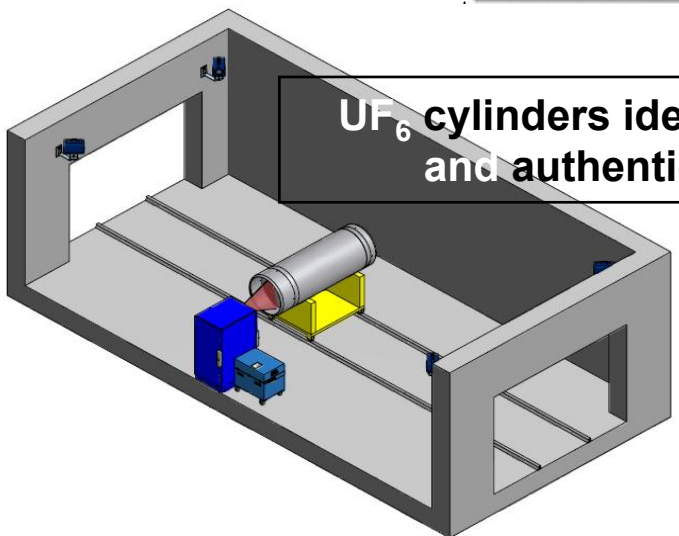
Real-time monitoring of load cells



Modelling of cascade



Improved NDA for enrichment verification in product cylinders



UF_6 cylinders identification and authentication

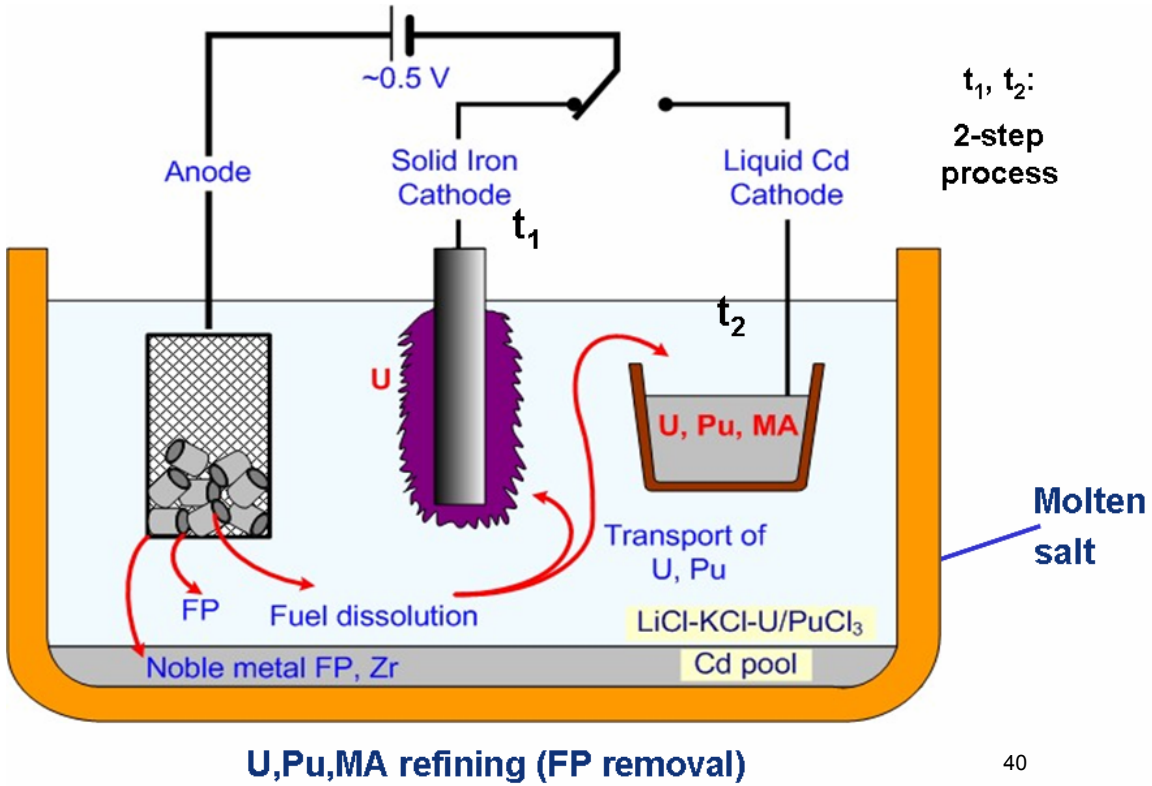
RFID cylinder tagging

Portable mass spectrometers

Research objectives and background:

Development of advanced partitioning methods, for example:

Electrorefining of metallic fuels



U,Pu,MA refining (FP removal)

40

➢ **Pyroprocessing**

Development of pyrochemical methods (salt melts) for the reprocessing of Actinides, also metal fuel cycle

► **NDA & modeling**

Particle Analysis

Analysis of uranium and plutonium in particles sampled in a broad range of nuclear safeguards contexts (environmental sampling)

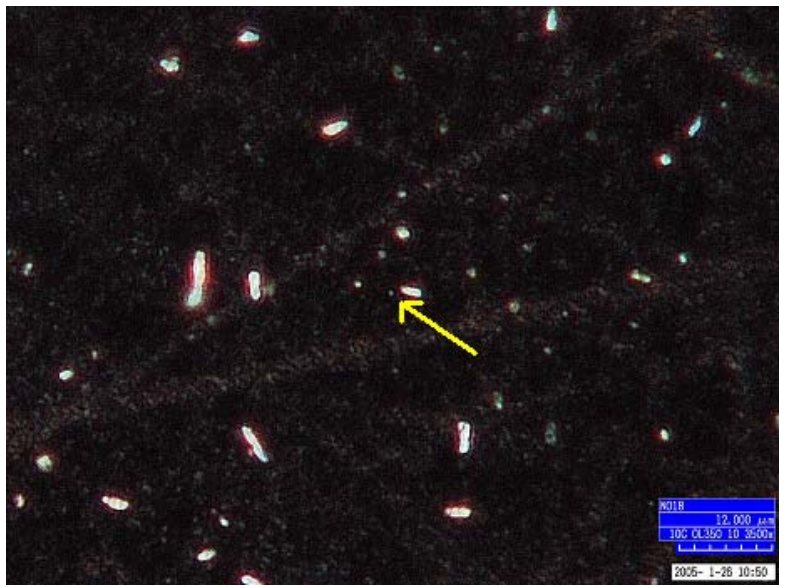
• Search through millions of particles to find the particles of interest.
A “needle in the “haystack” problem performed under strict time pressure.

• To make precise, accurate and timely measurements of both

major (U-235, U-238) and

minor (U-234, U-236) isotopes

(enrichment process, irradiated material)



Particle distribution (Resolution 1050x and 3500x)

→ **Need for increased mass resolution**

Particle Analysis

A prioritised R&D project at ITU has been in implementing **LG-SIMS** for uranium particle analysis to strengthen the abilities to detect undeclared nuclear material handling.

The installation of a LG-SIMS at ITU is planned for the **second half of 2011.**

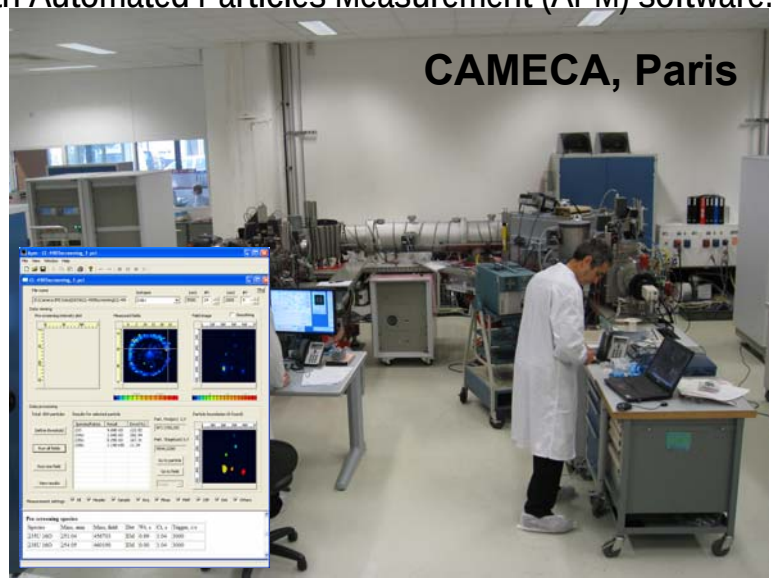
23rd of May: factory acceptance tests

The project is supported by DG-ENER, the main customer for the SIMS particle laboratory at ITU.

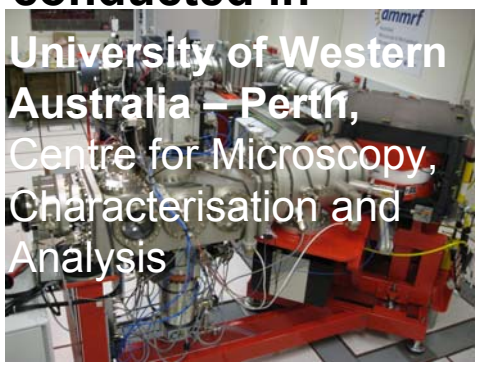
(DG-ENER finance one third of the costs)

IMS 1280 –

HR Secondary Ion Mass Spectrometer (SIMS) with Automated Particles Measurement (APM) software.

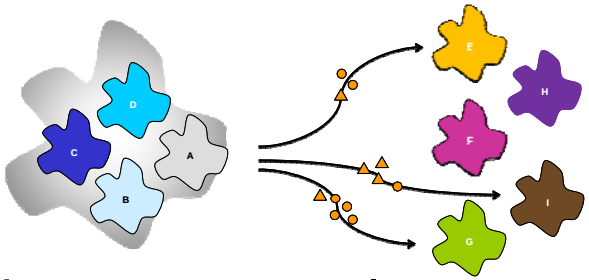


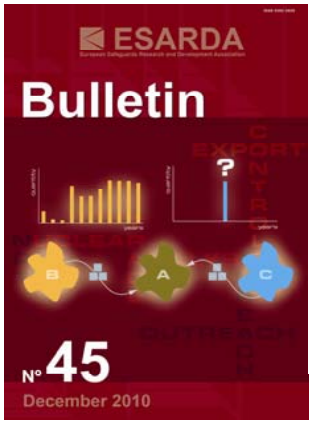
This work has been conducted in collaboration with:



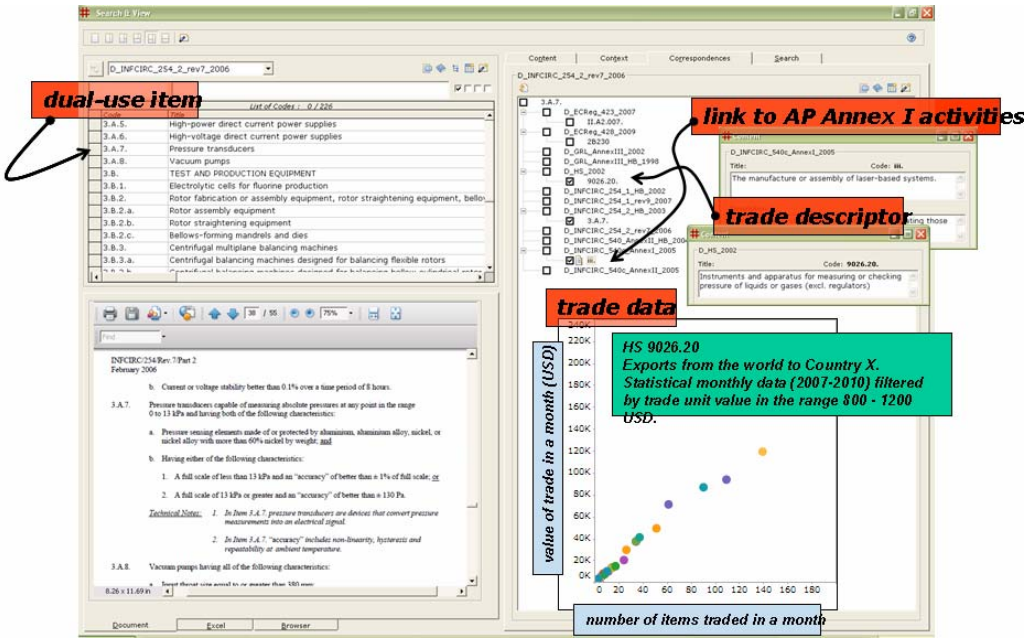
Analysis of Proliferation Concerns

- **Export Control Process**
  - Technical review
  - custom risk assessment
  - Support to policy implementation in export controls





- **Non-Proliferation studies**



- **Trade Analysis**
  - EC Support Programme to IAEA
  - Open source trade data
  - 'The Big Table': JRC tool in support to trade analysis Perspective: link with IAEA Physical Model
  - Estimate trade flows of dual-use / Identify trade deflections.
  - Monitor implementation of restrictive measures and catch-all controls.
  - Closer contact established in 2010 with DG TRADE and MS
  - Collaboration with US DoE

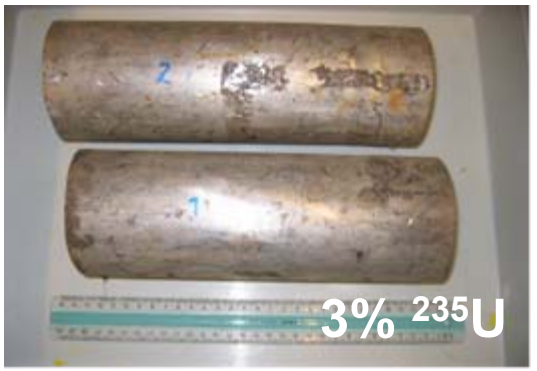
Nuclear Security: Illicit Trafficking / CBRN

- **Prevention**
  - ✓ Control over radioactive sources
  - ✓ Exchange of information (reporting; early warning)
  - ✓ Physical protection
- **Detection**
  - ✓ Development of advanced detection technologies
  - ✓ Intercomparison of nuclear detection techniques
- **Response**
  - ✓ Nuclear Forensics
  - ✓ National response plans

Cross-border cooperation,
International collaboration;
cooperation with third countries

Nuclear Security: Nuclear Forensics

- Some 10 cases of intercepted nuclear material subject to nuclear forensic analysis at ITU since last ESARDA symposium 2009
- Direct support to EU member states
- Application of analytical methods derived from safeguards analysis, material science and environmental analysis

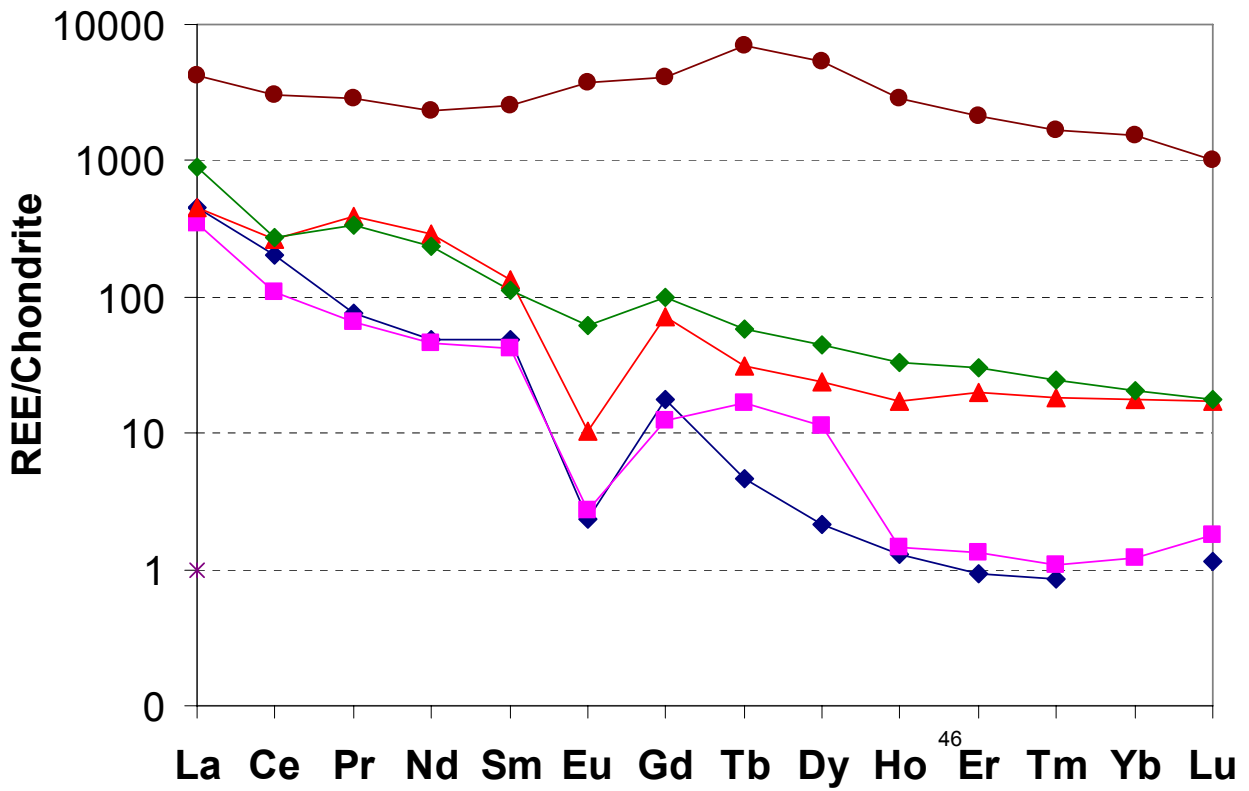


Nuclear Forensics: A case study

Haigerloch uranium cubes produced in 1943.

From which mine did the uranium come?

Jáchymov / Joachimsthal in the Czech Republic or Belgian Congo?



> 70 a

➔ **This is part of a systematic study on trace elements in U materials**

Training and Education

Establishing an EU Nuclear Security Training Centre at ITU for 2012

- With support of EC Directorate-General for Home Affairs (DG-HOME)
- Ensuring high standard in detection and response in EU-27 and beyond

- Complementary to national training activities
- Focus on advanced training using nuclear material
- In collaboration with the EU MS, the IAEA and international initiatives (e.g. BMWG, ITWG, GICNT)

Towards a JRC Training and Education integrated concept:

A European School in Nuclear Safety and Security

- Assemble related activities in JRC-Nuclear Safety and Security under a single heading
  - Information / Education / Training (in-house, external) /Knowledge management
- Key T&E Areas
  - Nuclear Safeguards, Security and Forensics
  - Nuclear Fuel Cycle, with emphasis on nuclear fuel and fuel processing
  - Basic Nuclear Science, with emphasis on the physics and chemistry of the actinides and on nuclear data

Competences of JRC are unique or strongly complementary to those of the academic institutions and other T&E initiatives

Summary & Perspectives

Nuclear Safeguards

- S/T support – operational and R&D – to Safeguards authorities DG ENER & IAEA
  - New nuclear materials (MOX measurements, e.g.)
  - Alternative-fuel-cycle accountancy (pyro-separations,…)
  - Deploy state-of-the-art instrumentation, competitiveness (LG-SIMS, e.g.)
  - Process monitoring
  - Enhanced detection of clandestine activities
  - Reference materials for absolute measurements

ESARDA

is an important platform and catalyser for exchanging views and recent developments in the safeguards field

mvm paks nuclear power plant

Capacity extension of Paks Nuclear Power Plant

ESARDA Safeguards Conference

16-20 May, 2011.
Budapest, Hungary

Géza Pekárik
technical director

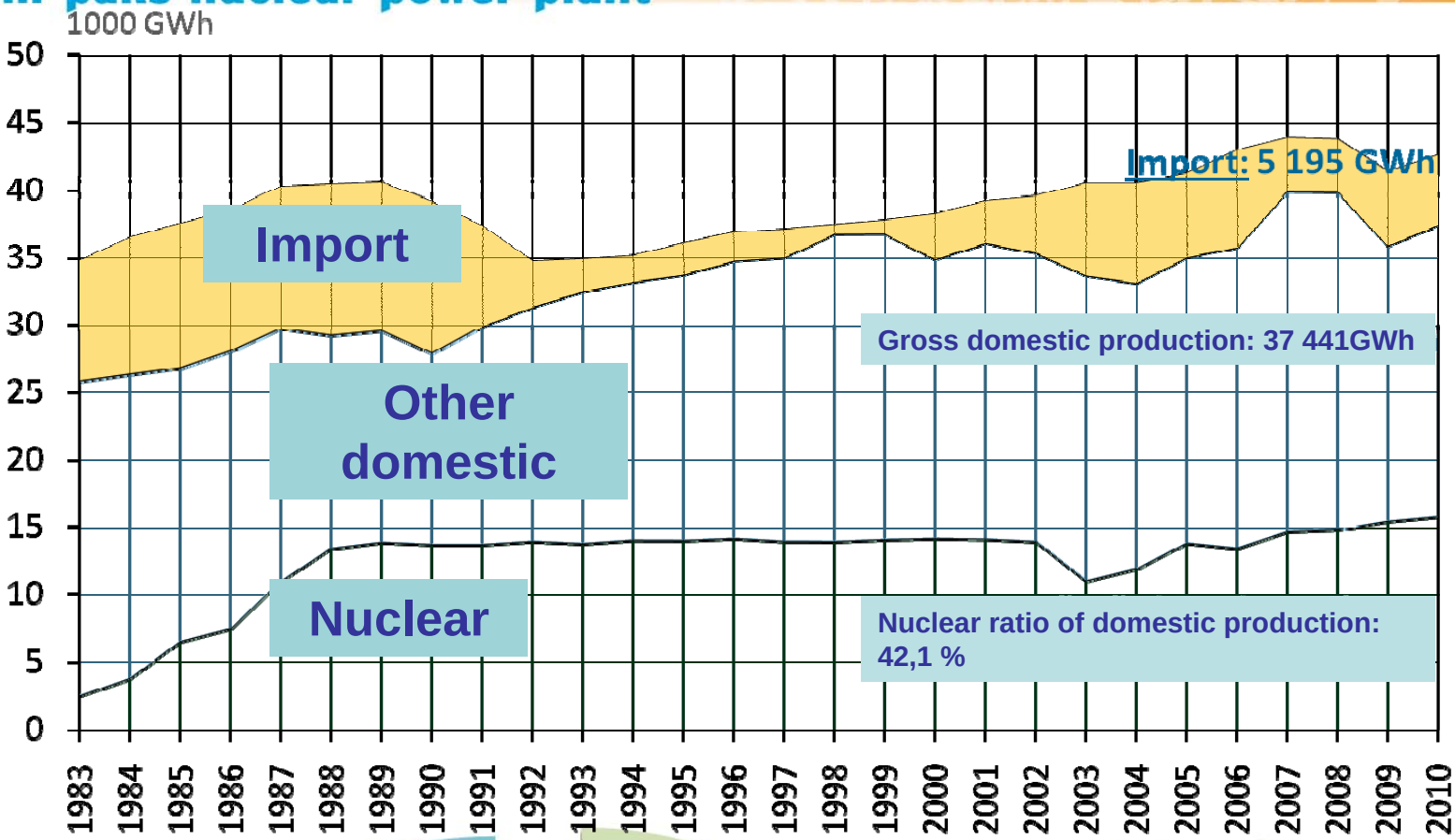
mvm paks nuclear power plant

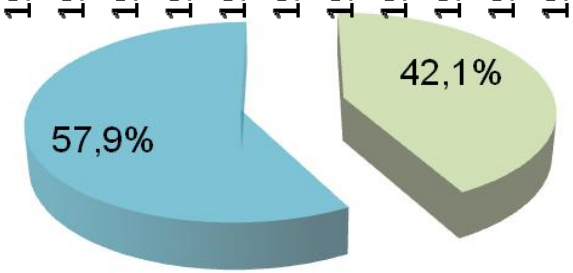
Plant overview

The four VVER-440/V213 units each with 440 MW electrical capacity were started in the years: 1982, 1984, 1986 and 1987

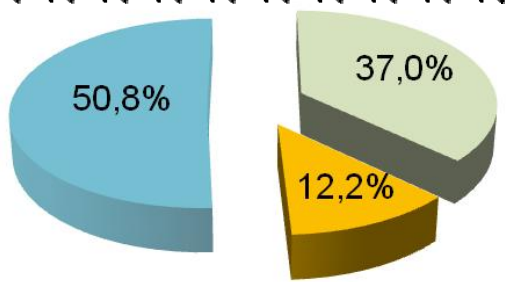
Hungarian Electricity Production

mvm paks nuclear power plant





Gross domestic production: 37 441 GWh

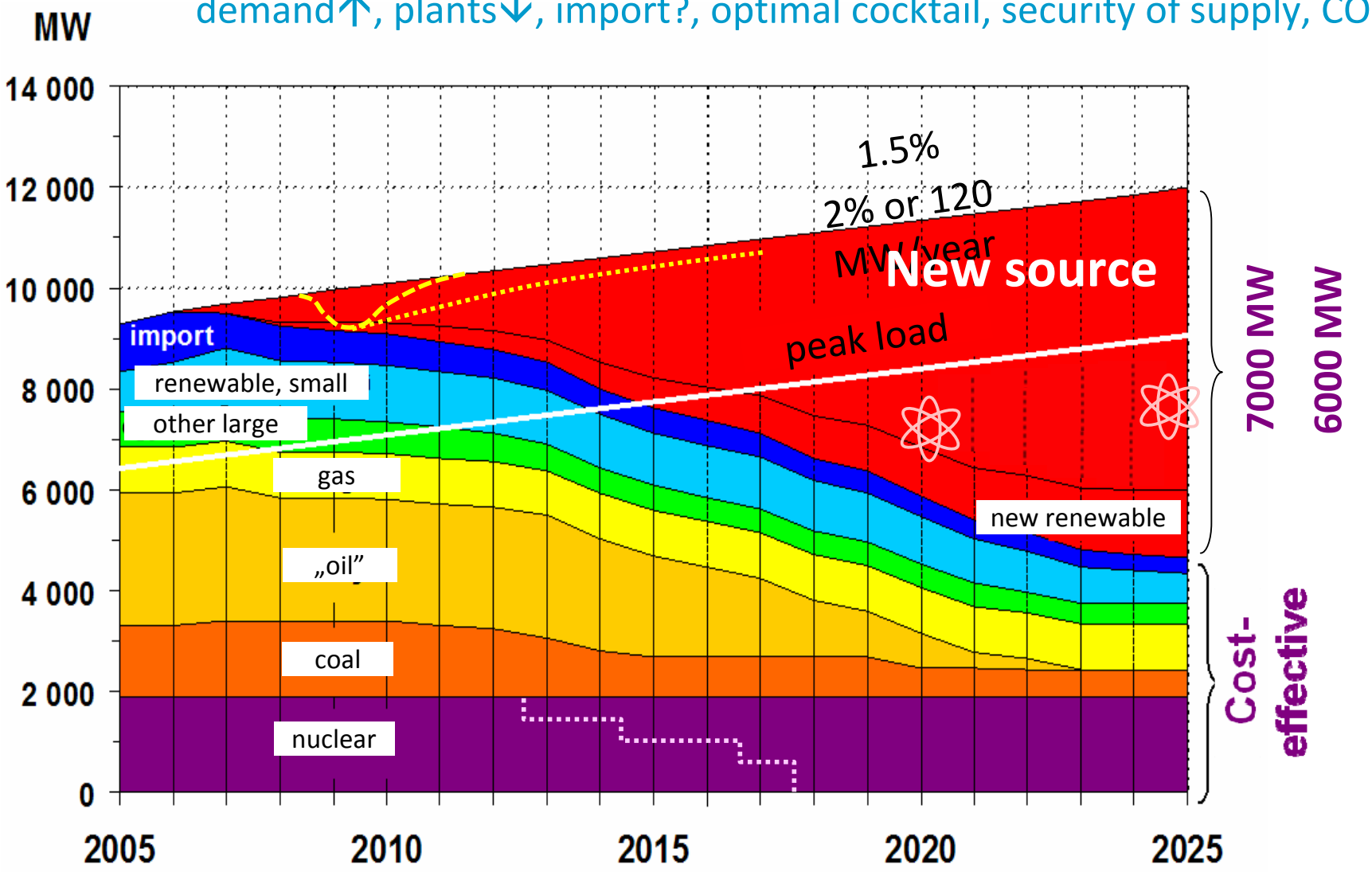


Gross domestic consumption: 42 636 GWh

52

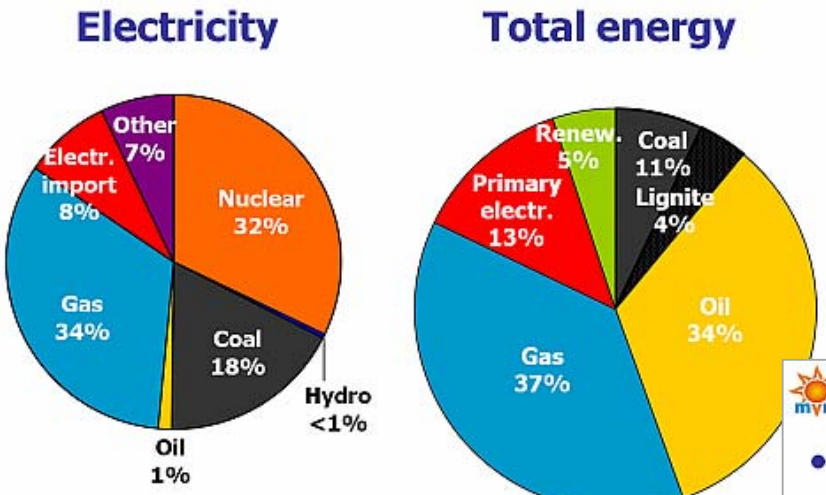
Domestic demand-supply

demand↑, plants↓, import?, optimal cocktail, security of supply, CO_2



Why nuclear? Sources and import

Source structures



HU, 2008

Net import dependency

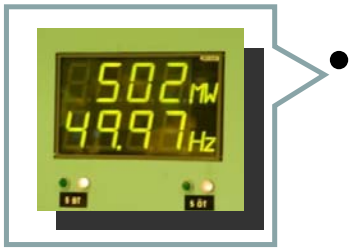
mvm paks nuclear power plant

• **Energy sources**	**[%]**	
– Nuclear fuel	100.0	
= from Russia:	100.0	(diversified)
– Gas	82.5	
= from Russia:	74.7	
– Oil	85.0	
– Coal	43.9	
– Electricity	11.3	
• **Import** (without nuclear fuel)	68.6	
• **Total import**	**79.4**	

HU, 2008

54

Nuclear capacity increase



- Power uprate (past)
  - the four Paks NPP units now on 108%
  - 440 ⇨ 460 ⇨ 500 MW

- Lifetime extension (present)
  - 30+20 years
  - licensing is in progress

- New nuclear build (future)
  - political support
  - preparation started

Preconditions, time scale

mvm paks nuclear power plant

7. § (2) For the start of preparation of new nuclear unit, or storage facility or extension of the existing NPPs by new unit is necessary to obtain the prior principal agreement of the Parliament.

Atomic Law

Teller project

2007	08	09	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25

years

studies

Teller studies, documents

- Topics studied
  1. Production, grid analysis
  2. Energy policy, strategy
  3. Economy, trade
  4. Public acceptance, communication
  5. Nuclear issues, environment
  6. Law and licensing
- Studies completed
  - Feasibility study
  - Preliminary environmental assessment
  - Analysis of the storage of spent fuel and radioactive waste from the new units

Fundamentals

- Power rate: grid and economy calculations
  - 600-1000-1600 MW
- Siting: Paks vs. other sites

quicker, cheaper at Paks!

- Not FOAK
- 60 years lifetime
- Light / pressurized water reactor (PWR)
- Load following capacity (50-100%)

G3 types (alphabetic order)

pressurized

AES-2006 VVER-1000 successor (Hidropress-Atomenergoproject, Russia)
AP1000 Advanced Pressurized Water Reactor (Westinghouse, USA)
APR-1400 Advanced Pressurized Reactor (KEPCO, S.Korea)
APWR1700 Advanced Pressurized Water Reactor (Mitsubishi, Japan)
ATMEA1 G3+ Pressurized Water Reactor (AREVA+Mitsubishi)
CPR-1000 Improved Chinese PWR (CNPEC, China)
EPR European Pressurized Water Reactor (AREVA, France-Germany)

boiling

ABWR Advanced Boiling Water Reactor (GE-Toshiba, USA-Japan)
ESBWR Economic Simplified Boiling Water Reactor (GE, USA)
SWR-1000 Siedewasser Reactor (AREVA+Siemens, German-French)

others

ACR-700,-1000 Advanced CANDU Reactors (AECL, Canada)
GT-MHR Gas Turbine Modular Helium Reactor (GA, USA)
PBMR Pebble Bed Modular Reactor (Eskom, USA-S.Africa)

Preconditions, time scale

7. § (2) For the start of preparation of new nuclear unit, or storage facility or extension of the existing NPPs by new unit is necessary to obtain the prior principal agreement of the Parliament.

Atomic Law

Parliament 19 March 2009

Lévai project

Paks-5

Paks-6

2007 08 09 10 11 12 13 14 15 16 17 18 19 20 21 22 23 24 25

years

licensing

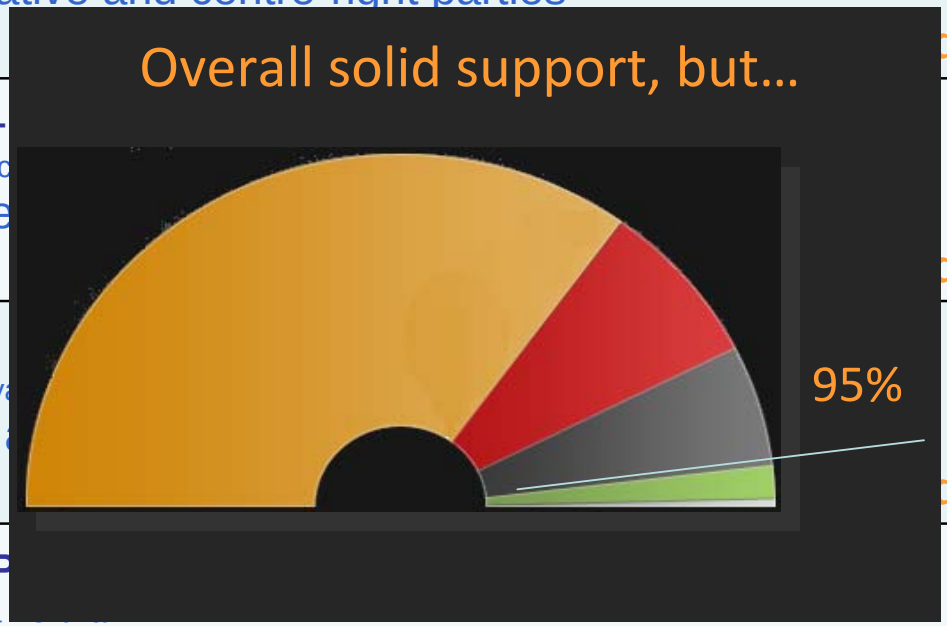
bid

design construction montage comissionig

60

Support in new Parliament

votes	share	party
263 (162)	**68%**	**Fidesz – Hungarian Civic Union** **KDNP – Christian Democratic People's Party** Fidesz Magyar Polgári Szövetség, Kereszténydemokrata Néppárt conservative and centre-right parties
59 (188)	**15%**	**MSzP –** Magyar Szoc social de
47 (0) NEW	**12%**	**Jobbik** Jobbik Magya populist
16 (0) NEW	**4%**	**LMP – P** Lehet Más a green liberal party for sustainable development and environmental protection, radical democrats **still no explicite statement on new nuclear build**
1		independent



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The Lévai* project

Lévai project

Enlargement of Paks NPP

Project Formation Document

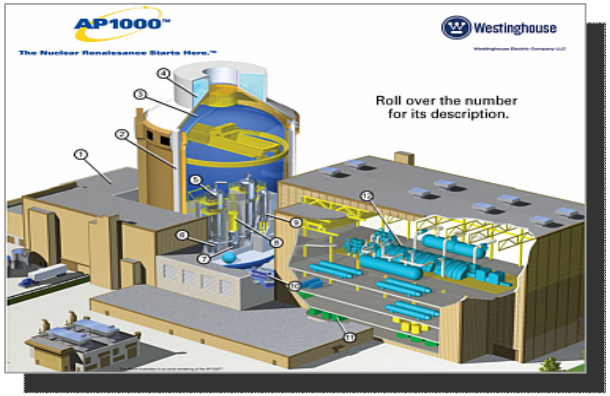
1 July 2009

- Established
  - in mid summer 2009
- Goal
  - define, start, and carry out in systematic manner all activities, by means could be rich the next milestone, the bid evaluation

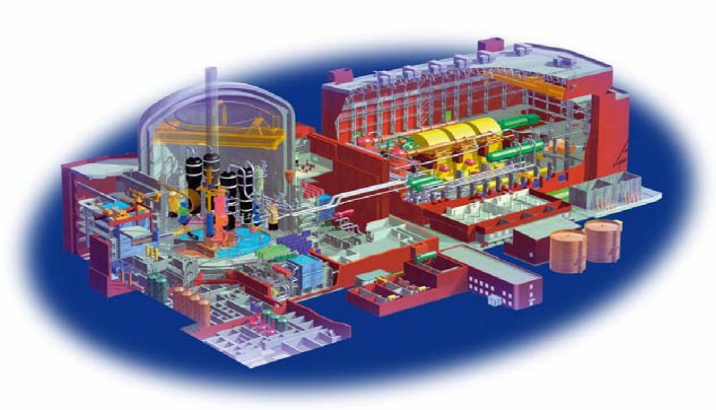
*prof. univ. András Lévai, 1908-2003

Designs for further consideration

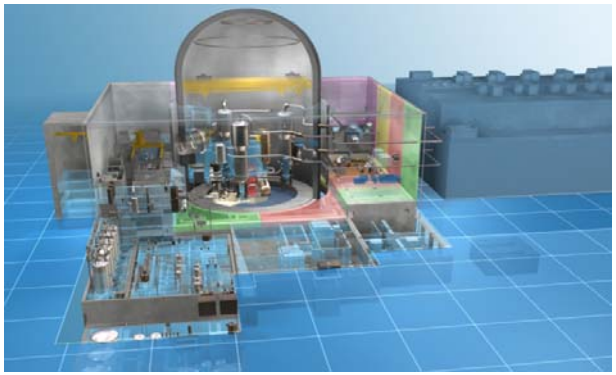
Westinghouse AP-1000



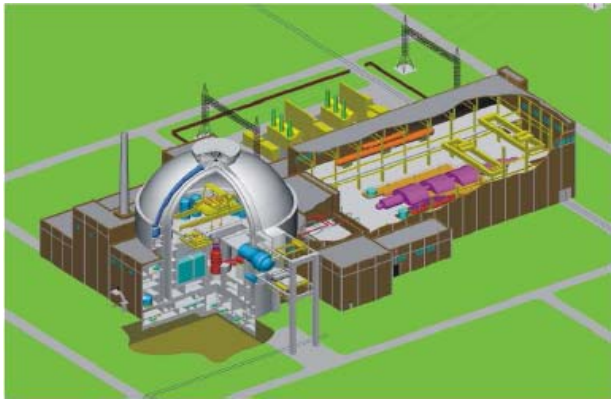
AREVA EPR



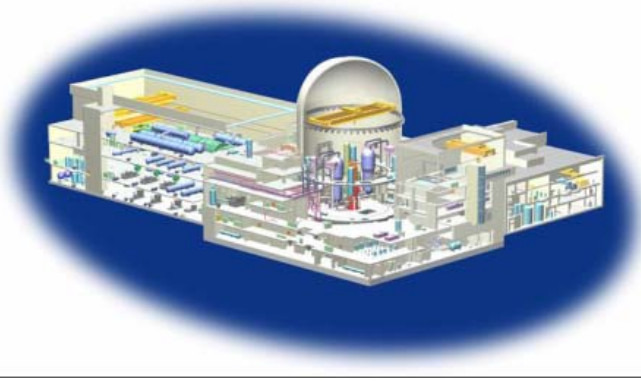
ATMAE ATMEA1



Atomstroyexport MIR1200



KEPCO APR1400



Financing

- **Volume of financing**
  - 12-15 years of continuous investment
  - investment ? (examples from World~ 3000 €/kW)
- **Own and investor sources**
- **Investors**

Strategic	Financial
– with NPP investment, operation experience, or – with design, fabrication, mounting capacity, or – electricity dealer, network company, or – base-load consumer	– commercial, development bank (domestic, foreign), or – export credit financing agency, or – international financial institution (EBRD, World Bank, EIB) – capital market (bond, …)

There are already 5-6 serious interested parties!

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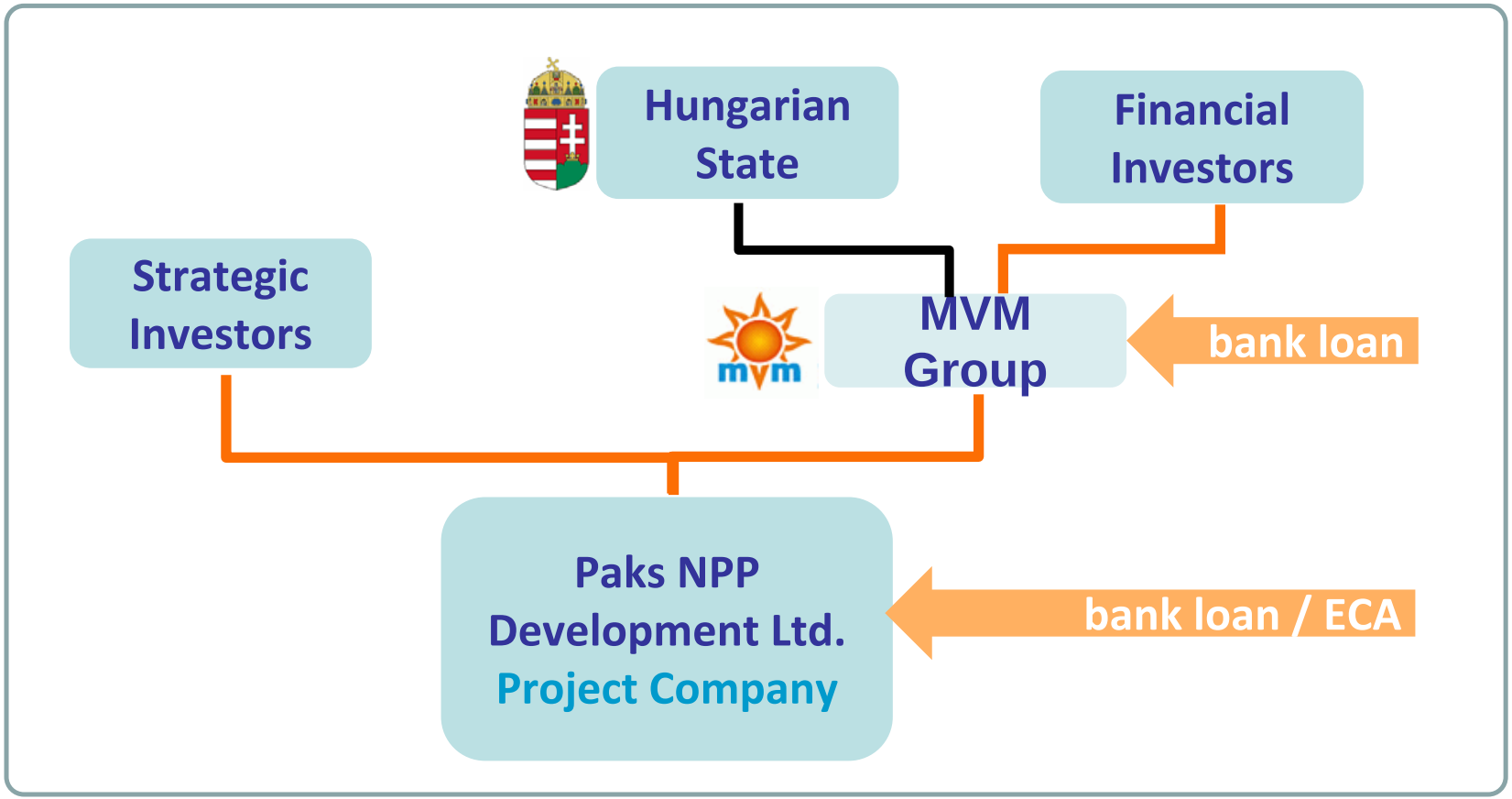
„In house" conception

- A proposed high-level financing model



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Bidding (technical issues)

• Possible examples of BIS content

Finnish ~ *
TVO (+Fortum)
FIN5 Bid invitation specification (BIS)
Helsinki, 2001-2002

Czech ~ *
CEZ,
Request for Information (RFI)
Prague, 2006

IAEA ~
Invitation and Evaluation of Bids for NPP,
IAEA-NG-T-3.9, Draft
Vienna, March 2010

* content known only on basis of detailed presentations

• Timing and need for human resources

Phase	Duration [months]	Number of experts
BIS preparation	4-12	20
Bid preparation	6-9	bidder staff
Bid evaluation	4-6	70 (15-20 full time)
Total for bidding	**14-27**	
Contracting	4-6	3-4 negotiators 10-15 supporting experts

In progress

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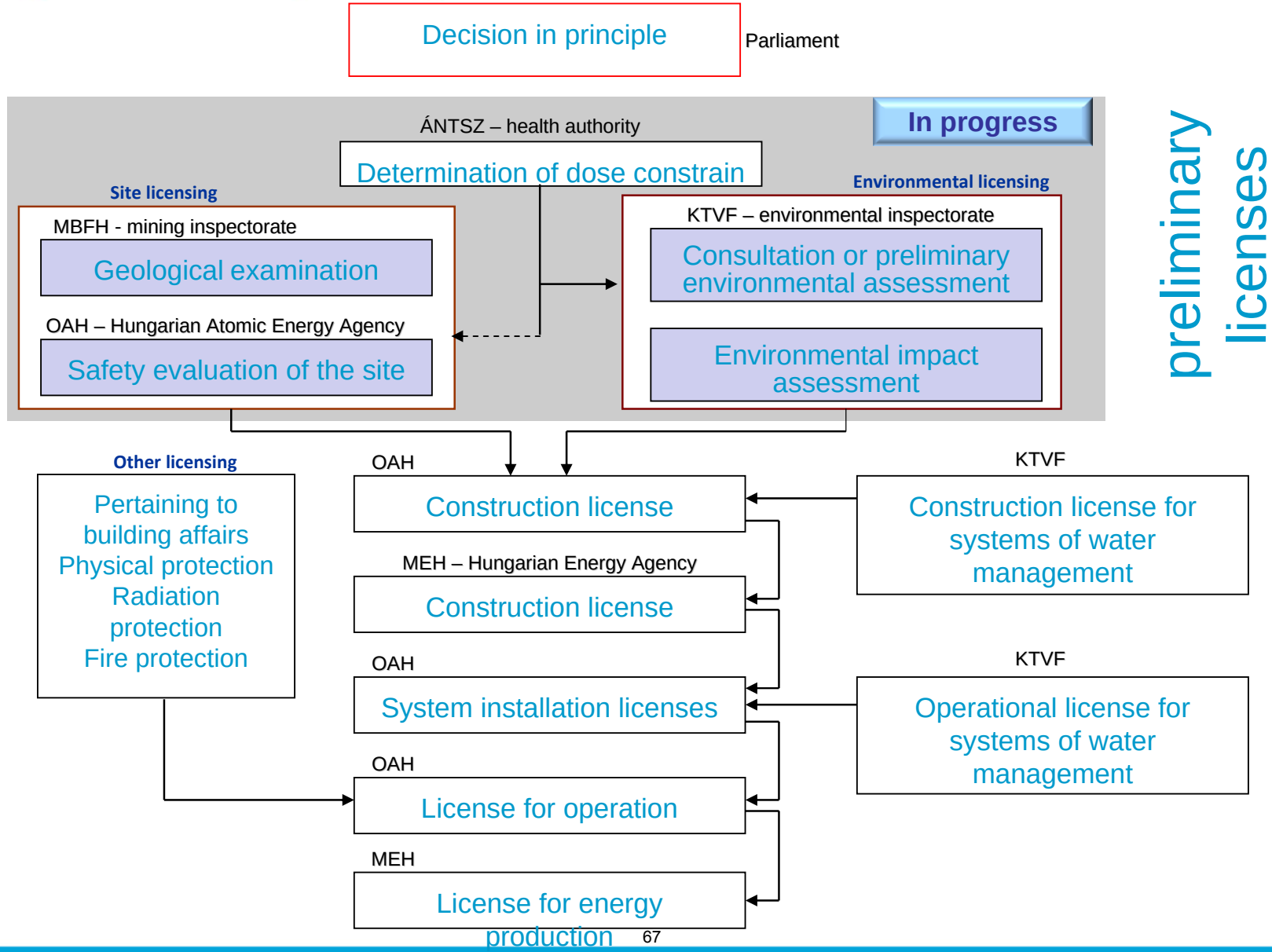
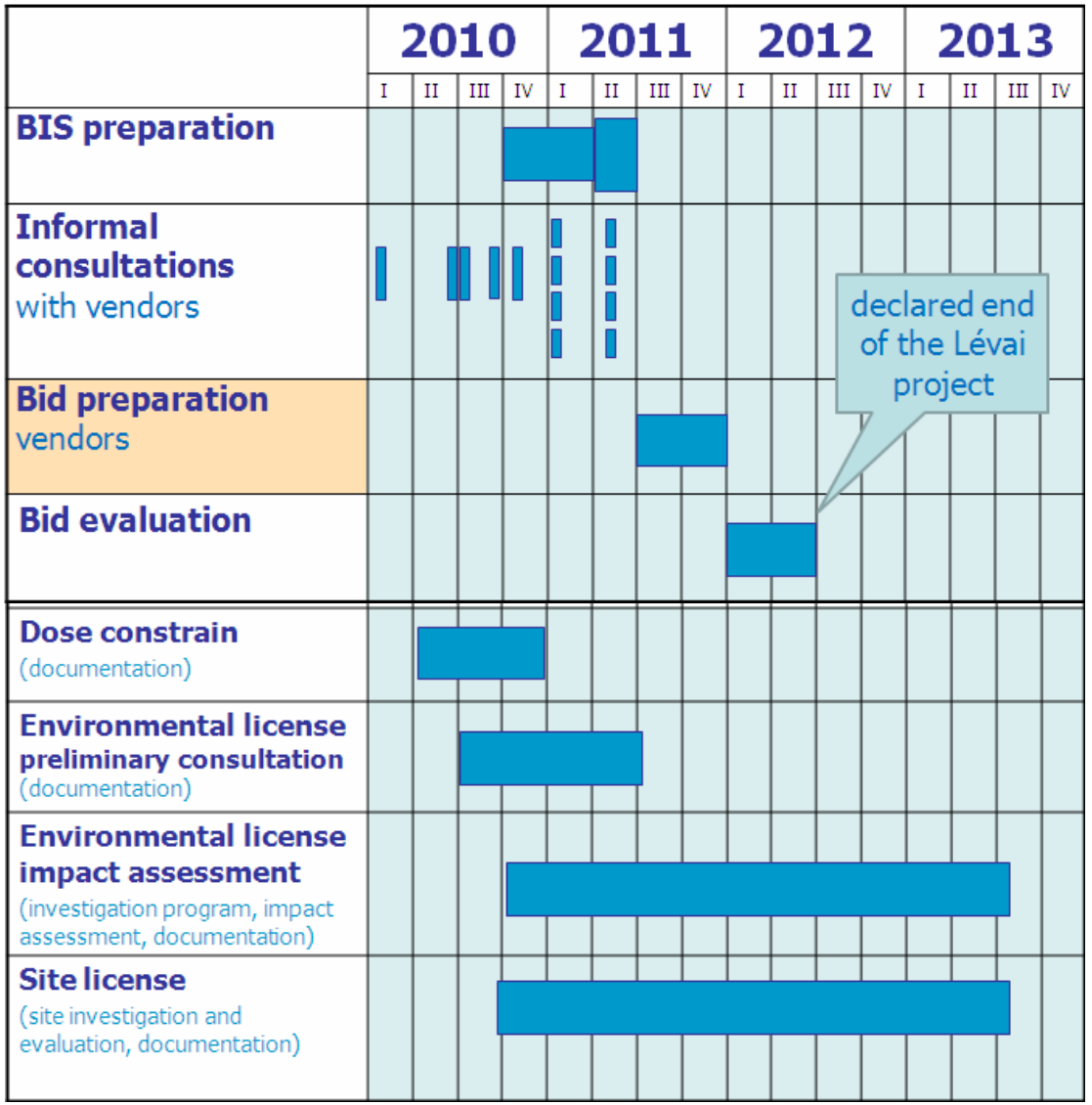
Licensing steps



General schedule

- **Bidding**
- **Licensing**



PR, communication

- **Organic continuation of the Teller project communication**

Communication

- **Organization: MVM & NPP committee**
- **Basic documents**
  - elaborated Q&A lists
    - = shorter, longer
  - strategic principles
    - = set-up and maintaining of a strategic partner network
    - = keep the new build in discourse
  - country wide surveys
    - = new build in the focus
  - publications
- **Other basic activities**
  - coontinous media presence
  - visitors at the NPP
  - professional conferences

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In progress

PR – past and present

- **New NPP website**
  - new build pages
- **Informative presentations**
- **Renewed Visitors' Center**
- **Exhibitions**
- **Guests**
- **Media**
  - regional press
  - commertional radios
  - natioal broadcast channels

New actions

- **MVM, NPP – Noguchi Portelli Novelli**
  - updated PR contract
- **Moving Visitors' Center**
  - exhibition on a road truck,
  - during half year: 100 towns/villages,

- **Two conference roadshows**
  - 5 city/7 city versions
- **Environmental licensing support**

prepare for normal/crisis communication

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Our vision

The view of Paks site shall be somehow like this at 2025:


mvm paks nuclear power plant
Thank you for your attention!

Synergy of safeguards, security and safety

Regulatory control

Dr. Kristóf Horváth
Head of Department
Department of Nuclear and Radioactive Materials
Hungarian Atomic Energy Authority

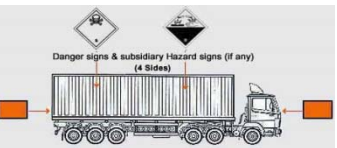
Introduction (1) – Fundamental objective

- The fundamental **safety-security-safeguards** objective of regulatory control:
  - **To protect people and environment**
  - **from harmful effects of *(any harm of)***
  - **ionizing radiation *(generated by the various applications of atomic energy)*.**
- without unduly limiting the operation of facilities or the conduct of activities.

Introduction (2) - Applications

- **Various applications of atomic energy (i.e. scope of regulatory control)**

  - Operation of nuclear facilities
  - Use, transport and storage of nuclear materials
  - Use, transport and storage of radioactive sources
  - Transport and storage of radioactive waste
  - Operation of equipment generating ionizing radiation (without a radioactive source)



Introduction (3) – Harmful effects

- **Harmful effects**
  - Personal exposures above limits
  - Environmental contamination
  - Environmental dose-rates, doses, activities, activity-concentrations above limits
  - Political, sociological, economical effects

Introduction (4) – Three ways

- **Ways how harmful effects may appear**
  - Incidents, accidents, severe accidents
  - Sabotage against nuclear facilities, nuclear materials, radioactive sources, and radioactive wastes
  - Malevolent use of nuclear materials, radioactive sources, and radioactive wastes *(or equipment generating ionizing radiation without sources)*

Goals of regulatory control

- To protect through
  - Prevention
    - Prevent deviations, harms
  - Detection
    - Detect deviations, harms asap
  - Response
    - Eliminate deviation, mitigate harm

Prevention

- Specification and official publication of regulatory requirements, continuous development of the requirement system in the light of the international experience, and scientific and technical evolution;
- Specification and official publication of enforcement procedures and penal acts;
- Licensing of activities in relation to establishment and operation of nuclear facilities and of various applications of nuclear and other radioactive materials;
- Registration of and accountancy for nuclear and radioactive materials and license holders;
- Establishment and maintenance of safety and security culture;
- Guidance and cooperation to support the meeting of the requirements.

Detection

- Inspection of compliance with preventive requirements;
- Environmental monitoring;
- Personal monitoring;
- Registration and accountancy verification;
- Operation of surveillance and detection systems;
- Evaluation of regular and occasional reports, and public information.

E.g. camera

- Safety (refueling machine)
  - Radiation resistant, operation and good picture under water,…
- Safeguards (surveillance of fuel movement)
  - Low light, remote connection, data storage, assessment of differences on photos, resolution, positioning…
- Security (assessment of alarms, detection)
  - Night and day, extreme weather conditions (fog, rain), high resolution, positioning

Response

- Operative measures:
  - Supervision of the implementation of measures defined in the procedures of the license holders;
  - Evaluation of event investigation reports, conduct of regulatory event investigations;
  - Conduct of an enforcement procedure.
- Joint actions:
  - On-site inspection and in-situ measurements;
  - Law enforcement and nuclear forensics investigations;
  - Search of lost nuclear and other radioactive material;
  - Safe transport of nuclear and other radioactive material.
- Emergency management:
  - Implementation of emergency response plans;
  - Recovery.

Legal and technical principles

Legal principles

- Justification (risk and benefit)
- Independence
- Provision of resources
- Safety-security-safeguards principle
- Optimization
- Responsibility
- Continuous supervision
- Protection of present and future generation (sustainable development)
- Liability/compensation
- Following technical and scientific developments
- Transparency/protection of sensitive information
- Predictability
- Graded approach
- Establishment of safety and security

Technical principles

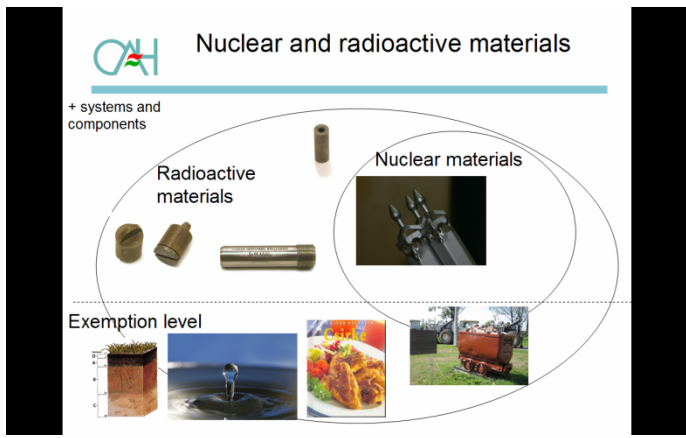
- ALARA principle
- Application of reliable technical solutions
- Deterministic approach
- Probabilistic approach
- Defense in depth
- Establishment of the design basis
- Use of negative feedbacks

Deterministic and risk-informed approaches

- Definition of design basis (accident, threat, Q(t))
- Definition of unacceptable consequences (dose limits, targets)

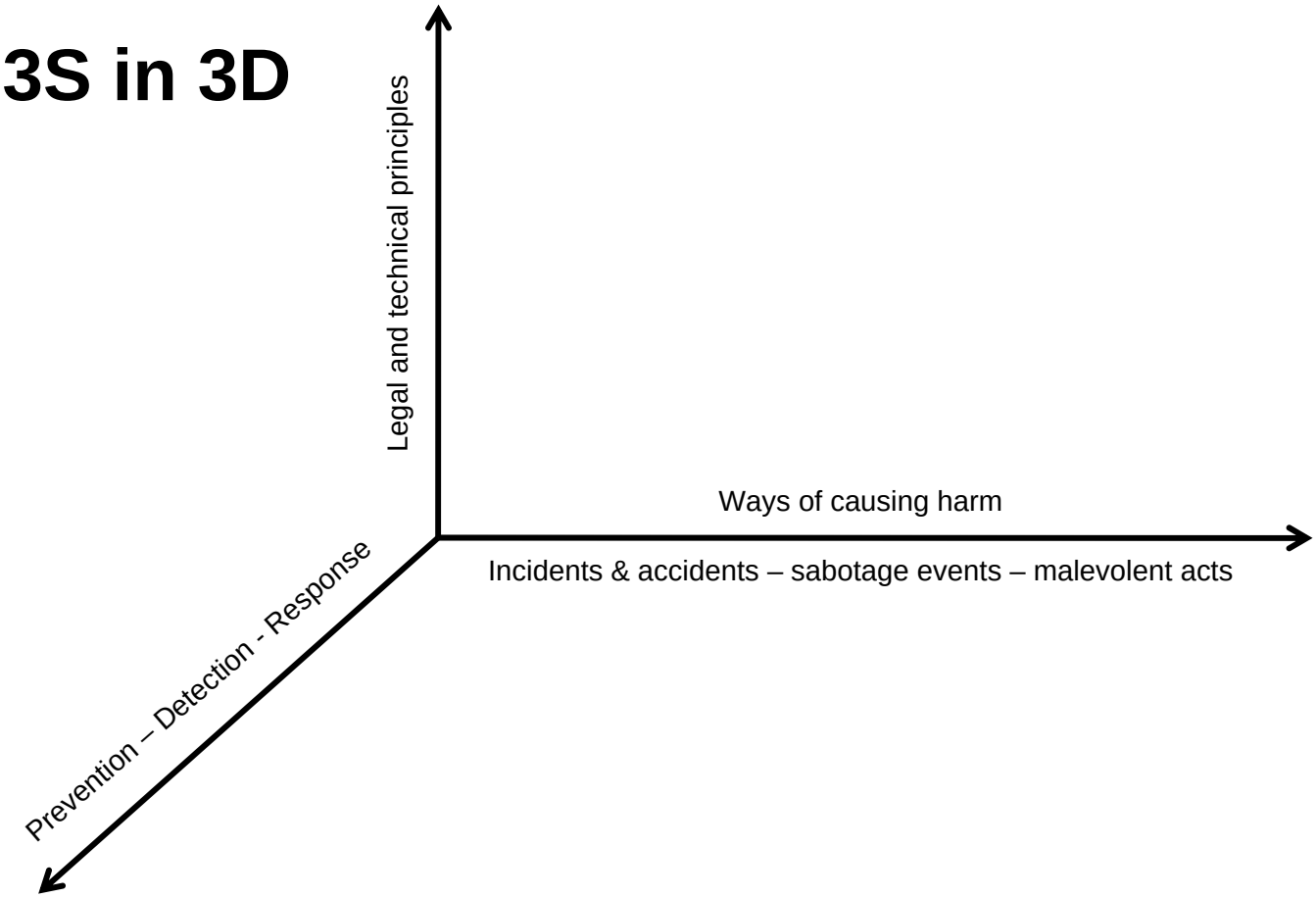
- Categorization of nuclear materials, radioactive sources, radioactive waste
- DSA and PSA

- Evaluation of users



Conclusion

Synergy: 3S in 3D



Köszönöm a megtisztelő figyelmüket!

Institute of Nuclear Materials Management

INMM

33rd ESARDA Annual Meeting

Symposium on Safeguards and Nuclear Material Management

INMM: Looking to the Future in Nuclear Materials Management

Ken B. Sorenson, Vice President
Institute of Nuclear Materials Management

May 17, 2011
Budapest, Hungary

Best practices organizations like INMM and ESARDA must be flexible and responsive to the dynamic world of nuclear materials management.

The evolving linkages between proliferation risks, nuclear capabilities in non-weapons states, and legitimate use of civilian nuclear power create an environment that requires constant assessment how we conduct our business.

How will we respond?

INMM
INSTITUTE OF NUCLEAR MATERIALS MANAGEMENT

ESARDA
European SAfeguards Research and Development Association

Both organizations have recently engaged in strategic planning efforts to better align their organizations with the dynamic world of nuclear materials management:

- **INMM – Strategic Planning Working Group on Organizational Structure**
- **ESARDA – Reflection Group 2010**

A core component of the INMM strategic planning was an Externality Analysis. Specific aspects considered in this analysis included:

International Oversight Activities

- IAEA — UN resolutions, IAEA documents and protocols
- G8 — Reaffirmation of NPT, FMCT, START
- Europe — Euratom, ESARDA, WINS

G8 Summit, July 2009
L'Aquila, Italy

Growth of Civilian Nuclear Power

- Asia — Strong growth in China, South Korea, India, Japan
- Middle East — Strong interest shown across the Middle East evidenced by the signing of numerous bi-lateral agreements
- U.S. — License applications and expressions of interest remains strong
  1 new enrichment facility on-line and 3 more licensing activities underway

U.S. Centric Issues

- Administration position
  - "…nuclear terrorism is the most immediate and extreme threat to global security." Quote from Mark Lippert, Chief of Staff of the National Security Council
  - Reduce and eliminate nuclear weapons
  - Ratify the CTBT
  - Begin negotiations on a FMCT
  - Secure all loose nuclear materials in the world within 4 years
  - Halt proliferation to new States
  - Negotiate/replace START
  - Strengthen the NPT
  - Global Nuclear Summit, Wash DC, March 2010
  - Cancel the Yucca Mountain spent fuel repository project

Obama Prague Speech
April 2009

- Department of Energy
  - Continue efforts on securing Nuclear Weapons Complex (material consolidation, PP, cyber-security, security forces)
  - Continue Global Threat Reduction Initiative
  - Construction of MOX facility in South Carolina
  - R&D on fuel cycle alternatives

Questions arising from these linkages

- **Non-proliferation and Commercial power production**
  - **How can the expansion of nuclear power proceed while minimizing the risks of technology and materials proliferation?**
  - **What roles do bilateral and international treaties have in managing this balance?**
  - **What roles do commercial organizations such as the Nuclear Suppliers Group have in minimizing these risks?**
  - **What roles do states have?**
- **Physical protection and commercial power production**
  - **How do we transfer traditional PP protocols from a weapons protection strategy paid for by governments to protection requirements paid for by private industry?**
  - **How do we integrate PP requirements across international boundaries as the fuel cycle becomes increasingly global?**

Externalities Analysis – Observations

- The concern over nuclear materials management is global
- There is a strong emphasis on treaty ratification and verification
- There is strong U.S. engagement to minimize proliferation risk while encouraging commercial expansion of nuclear power
- There are four major forces driving the future of nuclear materials management;
  - Concern over terrorism since 9/11
  - Dramatic reduction in the nuclear weapons stockpile
  - Concern over a growing number of countries acquiring sensitive nuclear materials/capabilities
  - Dramatic increase in commercial nuclear fuel cycle development
- The current INMM Technical Divisions seem to address the majority of these externalities
- However, changes are recommended to strengthen the overall Technical Division portfolio in light of these externalities, particularly as they relate to the commercial fuel cycle.

INMM Technical Division Structure

Old Structure	New Structure Based on Externalities Analysis
International Safeguards	International Safeguards
Materials, Control & Accountability	Materials, Control & Accountability
Non-proliferation and Arms Control	Non-proliferation and Arms Control
Physical Protection	Nuclear Security and Physical Protection
Packaging & Transportation	Packaging, Transportation, and Disposition
Waste Management	Facility Operations

Two new Standing Committees have also been established

- **Strategic Planning**
- **Education and Training**

INMM is committed to working closely with our international partners in the constant effort to improve best practices in nuclear materials management

International Atomic Energy Agency

- **INMM has Standing Observer Status at the IAEA General Conference**

World Institute for Nuclear Security

- **INMM serves as a technical advisor**
- **WINS is a sustaining member of INMM**
- **Joint WINS/INMM workshop on human reliability, Fall 2011, Sellafield**

International INMM Chapters

- **Japan**
- **Korea**
- **Morocco (New!)**
- **Obninsk**
- **Russian**
- **United Kingdom**
- **Ukraine**
- **Urals**
- **Vienna**

INMM's relationship with ESARDA is strong and broad reaching

Joint Meetings

- **Ispra 1984**
- **Arona 1996**
- **Albuquerque 1998**
- **Tokyo 2000**
- **Como 2003**
- **Santa Fe 2005**
- **Tokyo 2008**
- **Aix en Provence 2011**

Joint Training

- **Nuclear Non-proliferation and International Safeguards Training Workshop: July 2011, Palm Desert, CA**

INMM Annual Meetings

Conclusion

INMM looks forward to a lasting and growing partnership with ESARDA in working together to improve nuclear materials management in the world in order to facilitate continued safe and secure expansion of civilian nuclear power while minimizing proliferation and security risks.

INMM
INSTITUTE OF NUCLEAR MATERIALS MANAGEMENT

ESARDA
European SAfeguards Research and Development Association

Euratom safeguards, a regional safeguards system

Paul Meylemans, Maurizio Boella, Johan Dackner, Wilhelm Koehne, Piotr Szymanski, Alain Thomas

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Abstract:

With the signature in 1957 of the Treaty establishing the European Atomic Energy Community, abbreviated Euratom, the six founding countries of what is the European Union today, laid the basis for a strong system of nuclear material safeguards. The legal basis for the European system of safeguards is embedded in European law that itself is binding in its entirety and is directly applicable in all Member States. The Treaty charged the European Commission with the responsibility of the execution of safeguards duties and of assuring that obligations assumed by the Community under agreements concluded with third states or international organisations are complied with. For the Commission to be in a position to discharge itself of that responsibility, it was given wide ranging powers.

The keeping of operating records at the level of the users of nuclear material and the basis for a European system of nuclear material accountancy was already laid down in the Treaty and developed in detail in a Regulation. The first safeguards inspection by the Commission was carried out 50 years ago in April 1960. The Community system of safeguards based on accountancy verifications and on-site inspections was further developed over the years and has evolved into a supranational system of safeguards for the European Union.

When the Non-Proliferation Treaty and the corresponding Verification Agreements were signed by the Community, its Member States and the IAEA, the Community system of safeguards was seen as the first layer of safeguards of which the IAEA would make use to the extent possible. Cooperation in a collaborative spirit allows both inspectorates to carry out their inspection activities without unnecessary duplication of safeguards activities or creating any undue burden on the nuclear operators. The Euratom Community and several of its Member States have continuously supported the IAEA activities in the field as well as by means of research and development of instruments, tools and methods. More recently, the Euratom Community ensured that the IAEA was able to implement Integrated Safeguards in all European Union Member States that have nuclear activities.

Also today, the Verification Agreements remain a solid, legal basis for further collaboration, which the Euratom safeguards system is ready to develop with the IAEA for the additional tasks and challenges the two inspectorates will be facing in the next 10-20 years.

Keywords: Euratom; regional safeguards system, IAEA, cooperation

1. Introduction

The European Union has a well established regional safeguards system. That safeguards system finds its origin in the Treaty establishing the European Atomic Energy Community (Euratom). That Treaty is often referred to as the Euratom Treaty and its safeguards system as Euratom safeguards. The Euratom Treaty was signed in 1957 and entered into force in 1958. Euratom safeguards has a very strong legal basis that is binding for all Member States and has performed inspections since the early sixties.

Also in the mid fifties other countries and regions around the world wanted to make use of nuclear energy for the production of electricity. In the aftermath of World War II, there was a clear need to ensure that the nuclear materials needed for the production of electricity would not be diverted for the production of nuclear weapons. With that aim, the International Atomic Energy Agency (IAEA or the Agency) was established in 1957, but the Treaty on the Non-Proliferation of Nuclear Weapons, also known as the Non-Proliferation Treaty, was only opened for signature in 1968 and entered into force in 1970. The Non-Proliferation Treaty makes a distinction between the nuclear weapon states and the non-nuclear weapon states. Non-nuclear weapon states party to the Non-Proliferation Treaty undertake to accept IAEA safeguards on all source and special fissionable material in all peaceful nuclear activities, as defined in a Verification Agreement, also referred to as a comprehensive safeguards agreement, and its related Protocol. Nuclear weapon states party to the Non-Proliferation Treaty do not have a safeguards obligation, but have nevertheless concluded with the IAEA a Verification Agreement, that is referred to as a voluntary offer safeguards agreement, and a related Protocol.

All European Union Member States are party to the Non-Proliferation Treaty and all non-nuclear weapon states of the Union have entered into a comprehensive safeguards agreement with the IAEA. The United Kingdom and France, the two nuclear weapon states of the Union have concluded a voluntary offer safeguards agreement. The Euratom safeguards system has since the early days of IAEA safeguards been cooperating with the IAEA at many levels: by financial contributions, by providing advice and support, by undertaking joint inspections on the territory of the European Union Member States.

Events that happened since the early nineties, such as the discovery of undeclared nuclear activities in Iraq, the development of nuclear weapons by the Democratic People's Republic of Korea (North Korea) or the development of a uranium enrichment plant with components purchased from the black market by Iran, have lead the IAEA to broaden the scope of its responsibilities. These events made it clear that the Agency’s safeguards activities focussed on the verification of declared nuclear materials and declared nuclear activities were insufficient to detect undeclared activities or undeclared nuclear materials. This situation prompted the Member States to provide the Agency with additional rights and the Agency to rethink the implementation of its safeguards activities. In turn, this lead to the development of the Additional Protocol to the Verification Agreement, providing the Agency the right to make complementary accesses, which enable the detection of undeclared activities or undeclared nuclear materials at declared sites.

Historically, in the European Union, there is a strong culture of cooperation between the IAEA and the Euratom inspectorate with the aim of avoiding any undue burden to civil nuclear operators. The implementation of Integrated Safeguards by the IAEA for nearly 2 years now have lead to some issues which need to be resolved as a matter of some urgency. When doing that it may also be useful to investigate if and how the IAEA may profit from the work done by Euratom safeguards.

The question we want to address in this paper is: "How can cooperation be intensified to allow for a clear division of tasks between Euratom safeguards and IAEA safeguards, which optimises the use of resources, and continues to ensure that there is no unnecessary duplication of safeguards activities?"

2. Euratom safeguards

In 1957 six western European countries - Belgium, France, Germany, Italy, Luxembourg and The Netherlands – established a Treaty in which they undertook to share their efforts for the development of the peaceful use of nuclear energy and that would give them access to the technology, the know-

how and the source materials to benefit from that energy. However, in the post war period nuclear energy was quickly associated with the disastrous effects of nuclear weapons, as people had witnessed towards the end of World War II. That is why countries like the United States and the United Kingdom, two countries that could provide technology and source materials to make use of nuclear energy for the generation of electricity, requested serious guaranties that the materials provided would at all times be safeguarded and used for the declared peaceful purposes [1]. The Treaty, establishing the European Atomic Energy Community, therefore includes a powerful chapter on safeguards (Chapter 7 Safeguards, Articles 77 to 85).

Since its foundation in 1957, the European Union has developed into an important legal, economic and political entity, it has been confirmed legal personality with the entry into force of the Lisbon Treaty end 2009. The Euratom Treaty, mostly unchanged, is now an annex to the Lisbon Treaty and is equally applicable in all European Union Member States. Today the 27 Member States of the European Union are all part of Euratom and the civil nuclear materials on their territories are all subject to Euratom safeguards.

The Lisbon Treaty and the Euratom Treaty define a very strong legal framework. Both the treaties (primary law) and the regulations based on them (secondary law), are directly applicable in all European Union Member States, i.e. without the need to be transposed into national legislation in order to affect the citizens rights and obligations.

The Euratom Treaty charges the European Commission, the European Union's executive body, with the task to satisfy itself that in the territories of its Member States:

1. ores, source materials[1] and special fissile materials[2] are not diverted from their intended uses as declared by the users;
2. the provisions relating to supply and any particular safeguarding obligations assumed by the Community under an agreement concluded with a third State or an international organisation are complied with.

The European Commission, through its Directorate-General for Energy and in particular its Directorate for Nuclear Safeguards implements Euratom safeguards on the territory of the 27 Member States of the European Union.

The Euratom safeguards applies directly and primarily to the users or the undertakings holding or processing nuclear materials, but it also recognises the responsibility that a Member State has in ensuring the proper management of nuclear materials on its territory. The first Euratom safeguards inspection took place in 1960 and they have continued ever since, in all nuclear fuel cycle installations.

The Euratom Treaty also stipulates that the Community has the ownership of all special fissile materials. However, Member States, persons and undertakings have the unlimited right of use and consumption of special fissile materials which have properly come into their possession, subject to the obligations imposed on them by the Euratom Treaty, such as safeguards, and health and safety.

Euratom safeguards applies to ores, source materials and special fissile materials in the whole territory of the European Union, with the sole exception of materials intended to meet defence purposes. Contrary to the Non-Proliferation Treaty, the Euratom Treaty does not differentiate between nuclear weapon states and non-nuclear weapon states. That means that in the two nuclear weapon states in the European Union, i.e. France and the United Kingdom, all civil nuclear materials are also subject to Euratom safeguards.

The Treaty obliges persons or undertakings subject to safeguards to keep and produce operating records that allow for the accounting of ores, source materials and special fissile materials used or produced. These obligations are further detailed in Commission Regulation (Euratom) no 302/2005 on the application of Euratom safeguards.

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[1] Source material means uranium containing the mixture of isotopes occurring in nature; uranium whose content in uranium 235 is less than the normal; thorium.

[2] Special fissile material means plutonium 239; uranium 233; uranium enriched in uranium 235 or uranium 233

Although the Commission recruits its staff members from its Member States' citizens, these staff members are selected by means of an independent selection process. That selection process is designed to recruit candidates with the right mixture of general competencies and the required technical competencies for specific jobs such as nuclear inspectors. Commission staff members operate independently from their national governments and are only accountable to the Commission.

Staff recruited to be nuclear inspector go through a formal process of security clearance under the responsibility of the Commission's Security Directorate. Before the first assignment as a nuclear inspector, Member States are consulted if they accept the concerned Commission staff member as a nuclear inspector on their territory.

Nuclear inspectors have at all times access to all places and data and to all persons who deal with materials, equipment or installations subject to safeguards. In particular, nuclear inspectors have to obtain and to verify operating records. They immediately have to report any infringement they find to the Commission.

In the event of an infringement, the Commission may issue a directive calling upon the Member State concerned to take all necessary measures to bring the infringement to an end. In the event of an infringement on the part of the persons or undertakings subject to safeguards, the Commission may impose one of the following sanctions:

1. a warning;
2. the withdrawal of special benefits, such as financial or technical assistance;
3. the placing of the undertaking for a period not exceeding four months under the administration of a person or board appointed by common accord of the Commission and the State having jurisdiction over the undertaking;
4. total or partial withdrawal of source materials or special fissile materials.

This two-level system of sanctions – against the Member State or against the persons or undertakings concerned – allow the Commission to intervene directly, when required and at the most appropriate level. The political weight carried by even a simple warning is so high that even this seemingly light sanction is a very effective tool to achieve improvement.

As a general requirement, Member States have to ensure the fulfilment of all obligations imposed by the Treaty and to facilitate the achievement of the Community's tasks.

In summary, the Euratom Treaty defines a simple but very powerful system of safeguards that is based on a system of nuclear material accountancy and declarations made by users of all civil nuclear materials coupled to a system of inspections to verify that declarations are in conformity with reality. The Euratom safeguards system also has the required legal power to act directly upon users of nuclear materials who are not fulfilling their obligations.

3. Long standing cooperation with the IAEA

Since the early days of IAEA safeguards in Europe, the Euratom Community has cooperated with the IAEA. In 1973 all non-nuclear weapon states of the Community, the Community and the IAEA signed the Verification Agreement, also known as INFCIRC/193. The nuclear weapon states of the European Union, the Community and the IAEA signed separate tripartite Verification Agreements: for the United Kingdom it is the Verification Agreement known as INFCIRC/263 of 1976 and for France the Verification Agreement INFCIRC/290 of 1978.

Cooperation between the Euratom and IAEA inspectorates has been firmly built into the Verification Agreements. INFCIRC/193 specifies that safeguards activities under the Agreement are carried out jointly by the Agency and Euratom. Different from the model agreement INFCIRC/153, INFCIRC/193 has a protocol attached which is an integral part of the Agreement and amplifies certain provisions, especially on cooperation between the Community and the IAEA. The Safeguards Agreement and its Protocol indicate that the Agency shall make full use of the Euratom safeguards system (Agreement Art. 31). They specify that cooperation in the application of safeguards in the European Union aims at avoiding unnecessary duplication of Euratom safeguards activities (Agreement Preamble, Agreement

Art. 4, Protocol Art. 1). In determining the actual number, intensity, duration, timing and mode of the Agency inspections, account has to be taken of the inspection effort carried out by Euratom in the framework of its regional safeguards system (Protocol Art. 11).

The Safeguards Agreement and its Protocol therefore foresee that the Agency inspections are carried out during certain of the Euratom inspections (Protocol Art. 14). They charge Euratom with the general scheduling and planning of it's inspections in cooperation with the Agency (Protocol Art. 15) and specify that arrangements for the presence of Agency inspectors during the performance of certain of the Euratom inspections shall be agreed in advance between the Agency and Euratom (Protocol Art. 16).

The Protocol also defines that the Agency should work through observation of the inspection activities of the Euratom inspectors. For certain types of installations, the "observation regime" was later replaced in practice by a "joint team regime" under which Agency inspectors and Euratom inspectors were jointly performing inspection activities, with the aim of reducing effort on both sides. In 1992, the Director General of the IAEA Dr. Blix and Commissioner Cardoso e Cunha agreed to modify the approaches of observation and joint teams and to strengthen safeguards collaboration between Euratom and the IAEA with the aim to enhance the efficiency and effectiveness of safeguards implementation [2]. This concept became known as the “New Partnership Approach” and practical arrangements were developed under the guidance of the Liaison Committee.

European Union Member States also pay an important part of the IAEA regular budget. In 2011, all 27 European Union Member States together contributed more than 37% of the IAEA's regular budget. This is the largest contribution a comparable geographical area of Member States pays (USA 25%, Japan 12%, and the Russian Federation 1.5%). Several of the European Union Member States and the Euratom Community, through its Joint Research Centre, also contribute to the activities of the IAEA by means of their Support Programs.

4. Events that broadened the scope of the Agency’s mission

Since the seventies, the non-proliferation of nuclear weapons seemed to be well kept under control until the early nineties, when it was discovered that Iraq, a non-nuclear weapon state party to the Non-Proliferation Treaty, had begun developing a clandestine a nuclear weapons programme. To do so, Iraq had been using nuclear installations that were not declared to the IAEA. This discovery made it clear that the IAEA could no longer satisfy itself with the verification of declared activities but that it also had to develop its capability and explore possibilities to detect undeclared activities and undeclared nuclear materials.

Other major events demonstrating the limits of the IAEA safeguards system at that time include what happened in the Democratic People's Republic of Korea, in Iran and in Libya:

- The Democratic People's Republic of Korea ratified the Non-Proliferation Treaty in 1985, but gave notice of withdrawal from the Treaty in 2003 following U.S. allegations that it had started an illegal weapons program. In 2005 North Korea declared that it possessed nuclear weapons and executed a first nuclear test in 2006.
- Iran is party to the Non-Proliferation Treaty, but failed to declare for a certain period of time the existence of its uranium enrichment programme. The country has never been totally transparent about its intentions for the development of uranium enrichment. Critical components for its enrichment facility were purchased on the black market.
- Also Libya, while being a party to the Non-Proliferation Treaty and subject to IAEA safeguards inspections, decided to embark on a secret programme for the development of a nuclear weapon, in violation of its obligations. Libya was using nuclear material and technology provided by the A.Q. Khan network. Only when a shipment of centrifuge parts was intercepted, did Libya announce the elimination of its weapons of mass-destruction programme.

Also the events of 11 September 2001 demonstrated that major acts of aggression against a country can be committed by small but well organised groups of terrorists, not necessarily forming a state. It also became clear that such a terrorist threat could eventually involve the use of nuclear explosive devices or of nuclear “dirty” bombs.

During the 2010 Nuclear Security Summit, in which President Obama of the United States invited world leaders to Washington, it was recognised that nuclear security is a major issue for all countries around the world. The essential role of the IAEA in the international security framework was reaffirmed and the need for states to cooperate at bilateral, regional and multilateral levels was stressed. In the future it may also be expected that the IAEA's mission is expanded to activities related to the verification of nuclear disarmament.

Since the early nineties, the expectations of states around the world about the role of the IAEA in the area of nuclear safeguards and security have constantly grown. The IAEA is not only expected to safeguard declared nuclear materials and technology, but also discover undeclared activities and materials by a State and the illicit trafficking of nuclear materials by terrorist groups. Moreover, the IAEA may also be solicited to provide the necessary guaranties that, in the context of the dismantling of nuclear weapons, the recovered nuclear materials are consumed for peaceful purposes only. It is clear that each of these tasks is fundamentally different and requires specialised staff and methods. More fundamental is also the need for funding these different tasks and the resources required for it, which in a period of economic crisis is far from obvious.

Faced with these challenges the IAEA has reacted with different initiatives. In the nineties, the IAEA launched the idea of a strengthened safeguards system [3]. Strengthening measures included the availability of more information, increased access to facilities and other locations, and the enhanced use of advanced technology. These strengthening measures have been formalised in an Additional Protocol to the Verification Agreement. By signing the Additional Protocol, IAEA Member States provide the IAEA much wider possibilities than it had before.

In a state with only a comprehensive safeguards agreement in place, the Agency should be able to draw the credible conclusion that the declared nuclear material placed under safeguards remains in peaceful nuclear activities and is adequately accounted for. In a state with a comprehensive safeguards agreement and an Additional Protocol in force, the IAEA can draw the credible conclusion that **all** nuclear material in the state has been placed under safeguards and remains in peaceful nuclear activities and is adequately accounted for. In other words, the Additional Protocol provides the IAEA with the means to come to the credible conclusion about a state that there is no undeclared nuclear material or no undeclared nuclear activities and that declared nuclear facilities are only used for the declared peaceful purposes.

Reaching that conclusion requires the IAEA to perform a state-level evaluation of all information acquired in implementing comprehensive safeguards agreements and Additional Protocols as well as information available from other sources. The evaluation must show not only that there are no indicators of the diversion of nuclear material placed under safeguards but also that there are no indicators of the presence of undeclared nuclear material or activities in the state. The integration of the traditional safeguards measures with the strengthening measures has become known as Integrated Safeguards.

An important aspect of Integrated Safeguards is the IAEA's right to physical access. Both unannounced inspections and complementary accesses play an important role in drawing and maintaining the safeguards conclusions. In the European Union, where the IAEA had a long tradition of cooperating closely with Euratom for the execution of its inspections, unannounced inspections and complementary accesses have presented some implementation challenges. Due to a number of technical solutions that were already in place before the introduction of Integrated Safeguards, Euratom has managed to limit the number of unannounced inspections in the European Union and replace most of them with short notice random inspections.

5. Implementation of Integrated Safeguards activities

The implementation of Integrated Safeguards activities was recognised by the IAEA to be no small task. The number of complementary accesses that the Agency had to organise in states with an Additional Protocol in force would require extra resources, but the Agency was from the beginning aware of the fact that they had to achieve cost neutrality while maintaining quality and credibility.

The IAEA therefore had to find more economic but nonetheless credible and effective ways to implement the traditional safeguards activities taking due account of the additional assurance the IAEA has from its Additional Protocol-related activities. The introduction of unannounced inspections had to contribute to that goal. Unannounced inspections contribute to the detection of diversion of declared nuclear materials or misuse of a facility and are a deterrent to the use of declared material and facilities for undeclared activities. At first sight they also offer the prospect of cost-effectiveness. Through their unpredictability the Agency thought that they could replace more complex and expensive safeguards approaches. In other words, the main purpose of unannounced inspections was to be a cost-effective replacement for its traditional safeguards activities and to allow room for the organisation of complementary accesses that mainly focus on the detection of undeclared nuclear material and activities at sites and other locations. However the devil is in the detail.

In the European Union there is a long history of jointly planned and executed safeguards inspections based on the use of common methods and common equipment. Special arrangements have been developed over the years to ensure that the data obtained during inspections is shared between the two inspectorates and according to Article 21 of the Protocol to the Verification Agreement INFCIRC/193; the IAEA has always been kept informed about the results of Euratom inspections in which Agency inspectors did not participate.

The prospect of unannounced inspections triggered by the IAEA created serious concerns for the European Commission [4], who is in charge of organising safeguards inspections on the territory of the European Union, and also for a number of Member States that have restrictions embedded in their national law, imposing that IAEA inspectors are to be accompanied by Euratom inspectors. The Protocol to INFCIRC/193 interpreted unannounced inspections, as not announced to the operator but coordinated with Euratom, and thus no mechanism of information exchange was foreseen if the IAEA does an inspection alone, as this was legally not foreseen.

The execution of certain inspections by either Euratom inspectors alone or IAEA inspectors alone also raised questions related to the use of equipment and the detachment and placing of seals that had previously been carried out jointly. Activities that require both inspectorates to be present, but that have to be performed for operational reasons at a moment when only one of the two inspectorates is present, have created serious dilemmas for inspectors faced with this type of challenge in the field.

The concept of carrying out, as a complementary measure, a portion of routine inspections in an unannounced way was already permitted under the comprehensive safeguards agreement by Article 84. However they were subject to a number of conditions:

- both inspectorates have to coordinate all inspections with each other, including those unannounced to the operator;
- the Agency has to take into account the operator's operational programme;
- the Agency has to advise the Community and the state concerned periodically of its general programme of announced and unannounced inspections,
- the Agency has to make every effort to minimize any practical difficulties for the Community and the state concerned and for facility operators.

The changes to IAEA inspections implemented over the last couple of years underline that there are more and more differences between IAEA safeguards and Euratom safeguards. The major objective of Euratom safeguards as defined in the Euratom Treaty remains the detection of diversion of nuclear materials from declared peaceful activities. The IAEA has been making great efforts to redirect resources from its traditional safeguards activities to the activities required under the Additional Protocol, such as complementary accesses. Despite the clear requirement of the Verification Agreement to make full use of the Euratom system of safeguards and to avoid unnecessary duplication of Euratom safeguards activities, the Agency has decided to follow the less obvious path of triggering and performing inspections that are unannounced to Euratom or notified only at short notice, making effective and efficient use of resources more theory than practice.

6. Further enhancing Euratom-IAEA cooperation

This section is an outlook, an exploration of ideas, probably not comprehensive and not so much near term reality. Its intention therefore is to stimulate discussion and the exchange of ideas.

Already in 1993 [5] and again in the late nineties [6], Euratom argued for an intensified cooperation between the IAEA and Regional Systems of Accounting for and Control. The arguments to do so are clear and most of them still hold today.

6.1. How can Euratom further assist the IAEA?

In countries with undisputed high non-proliferation credentials, it is felt that the IAEA could decentralise a number of safeguards measures by relying more on the technical capability and the resources that can be provided by regional or national safeguards systems without, however, delegating its non-proliferation safeguards responsibility. In the context of an expanding mission, the IAEA may face difficulties to take equally care of verifications, information analyses and detection of undeclared activities and materials, tasks that also require different methods, competencies and tools. The decentralisation of certain safeguards activities to state or regional systems in a balanced way therefore appears to offer considerable advantages concerning the allocation of effort and appears to strengthen the operational role of the IAEA.

One way to delimit the safeguards tasks in the European Union between the IAEA and Euratom, could be to think of Euratom as the organisation to control and verify the “declared” use of nuclear material and the “declared” nuclear installations. That corresponds closely with the objective of Euratom safeguards as laid down in the Euratom Treaty. The European Commission continues to consider nuclear material accountancy and the control and verification of all declared nuclear materials in the whole nuclear fuel cycle of the European Union as a fundamental activity to guarantee the secure, safe and reliable operation of the civil nuclear facilities. It is a major activity to which it has dedicated an important amount of resources. Euratom provides the IAEA with accountancy declarations as well as the results of its safeguards inspections as required by Article 21 of the Protocol and the IAEA can make clear and full use of the Euratom system of safeguards as stated by Article 31 of the Verification Agreement.

Saying that Euratom would control and verify the declared use of nuclear materials and the declared nuclear installations, would not necessarily prevent the IAEA from performing its own verification activities as well, but it would at least allow the IAEA to concentrate on what currently seems to be the more important aspects of its objectives: the search for and detection of “undeclared” nuclear material, activities installations and sites.

The IAEA may develop guidelines for entering into a strengthened cooperation agreement with a supporting organisation. Of course, the IAEA should also be allowed to verify whether its guidelines are continuously met once such a cooperation agreement would be in place. This may take the form of system audits or unannounced participation in any of the inspections or activities carried out by the Regional or State System of Accounting for and Control.

6.2. Prerequisites for effective and intensified cooperation

The prerequisite for an effective cooperation is that the concerned parties actually trust each other, engage together towards common objectives and do what they promise to do. This may seem obvious, and it probably is when it comes to entering cooperation arrangements of a politically less controversial nature. But when it comes to issues such as nuclear material security, opinions differ on whether or not it is a wise course of action to trust the commitments of other parties and enter into a comprehensive cooperation.

Independency, therefore, has always been one of the cornerstones of the IAEA’s verification policies. The Agency should not give up its independency, but should realise that there are several ways to ensure that information provided by a cooperating organisation is of the expected quality. In the past, the IAEA has made use of advanced technology to ensure that data is genuine, such as authentication. However, such technical solutions may not always be applied. The fact that the IAEA has sufficient expert knowledge about the concerned facilities and has access to data originating from different sources gives the Agency the possibility to confirm the information provided by a cooperating organisation. Of course, the IAEA would at all times keep the right to verify a portion of the data it would obtain through a third party.

While not wishing to imply that the IAEA should delegate its safeguards responsibilities to others nor that it should abandon its rights to draw independent safeguards conclusions, it is felt that the IAEA could, technically, assign tasks with quasi-zero probability of diversion to a fully established regional or national system thereby justifying operational decentralisation, i.e. to reduce its inspection effort to a level required for quality control purposes only. At the same time it would allow the IAEA to concentrate on activities that for the moment seem to rise in importance, such as the search for and the detection of undeclared activities and materials and the illegal networks of proliferation.

6.3. Which criteria make Euratom a credible and capable partner to the IAEA?

Euratom is a Regional System of Accounting for and Control in charge of safeguarding the nuclear materials in the 27 Member States of the European Union. As a Regional System of Accounting for and Control, Euratom is not subject to the authority of any single state and can in that respect offer the IAEA far superior guarantees of independence compared to state systems for accounting of and control.

Euratom is based on a constitutional framework of political cooperation that exists between the Member States of the European Union and in which these States commit to non-proliferation. The legal framework operates on the basis of law that is directly applicable in each of the Member States. There is a court – the European Court of Justice – to which parties can appeal in the case of dispute and whose final decision is binding on the parties. Euratom safeguards resources are provided by the European Union within an established and transparent budget and financial framework, scrutinised by the European Court of Auditors. As a regional safeguards system, Euratom, has a proven record of safeguards effectiveness witnessed by the IAEA over several decades of close cooperation and inspection together in the field. The Euratom system is moreover under the supervision of the European Parliament and Council, which adds further elements of accountability and transparency.

Euratom has been performing safeguards inspections over the last 50 years and has gained important experience in safeguarding nuclear materials in all nuclear fuel cycle installations: conversion of ores and concentration, enrichment, fuel fabrication, power production, reprocessing, intermediate and long time storage of irradiated fuel and waste processing and final storage. In most of these installations and for many decades, inspections were carried out together with the IAEA, whose inspectors appreciate and recognise the quality of the Euratom safeguards inspections.

7. Conclusion

Since the introduction of Integrated Safeguards the European Union has been challenged with new safeguards implementation requirements. The two safeguards inspectorates still seem to be searching for a smooth integration of their activities and it is felt that there is still room for much closer cooperation which at the same time would be more in-line with both the Euratom objectives and the Verification Agreements and the IAEA's search to optimise resources to cover its enlarged responsibilities. With creative thinking, trust and goodwill, it must be possible to maintain credible and aligned safeguards inspections in the European Union — the fundamental idea of the Verification Agreement.

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"Making ESARDA Ready to Deal with the New and Future Verification Challenges"

ESARDA REFLECTION GROUP 2010 (draft) Report

ESARDA 33rd Annual Meeting

Budapest, May 16th 2011

M. Richard/CEA
On behalf of the Reflexion Group

PLAN of the Report

Introduction

1. Background and Present Status of ESARDA
   - History, foundation of ESARDA
   - Objectives of ESARDA
   - Current status of ESARDA
2. Identification of needs and Challenges
   - Establishment of a new reflection group RG 2010
   - ESARDA's forthcoming challenges
3. Coping to Identified Challenges
   - New framework: a broader role for ESARDA
   - The evolving nuclear fuel cycle: ESARDA's contribution
   - Reshaping ESARDA's R&D activities
   - Meeting ESARDA's members needs
4. Improvement of objectives, scope and management of ESARDA

Conclusion

STATUS of the REPORT

The RG 2010 report presents *the analysis and recommendations* of the 2010 Reflection Group

The need to keep the memory of the history of ESARDA, main lines of actions and events of the Association since its creation in 1969 as emerged during the drafting of the report which accommodate this need.

The target is:

⇨ ***to issue the report in July as some work is still needed***

⇨ ***to publish it, pending the agreement of the Steering as a special issues of the ESARDA Bulletin***

Termes of references

- **Terms of Reference of the 2010 Reflection Group agreed by the Executive Board/Steering Committee:**
  - ***to review the status of implementation of the decisions taken after the proposals of the RG 2000***
  - ***to assess the international and European context and trends in nuclear non proliferation and safeguards, security and disarmament verification areas and there impact on ESARDA Research and Development activities***
  - ***to analyze whether further actions and activities are needed in order to meet ESARDA members needs at European and international level***
  - ***to make proposals to the Steering Committee regarding possible improvements in the objectives, scope, structure and operations/management of the Association.***

Presentations

RG2010 work has been inspired and based on the following presentations:

Topics and presentations given during ESARDA 2010 Luxembourg:

- Export Control issues
- Nuclear Security Issues and the Fight Against Illicit Trafficking
- Comprehensive Test Ban Treaty update
- Modern Safeguards' Dependence on Information Technologies
- Fissile Material Cut-off Treaty

Presentations and papers given during RG2010

- ESARDA Reflection Group 2000: Objectives, Achievements, Recommendations
- Informal notes by Bruno Pellaud
- Nuclear forensic presentation
- Environmental analysis:
- EU MS Support Programme to IAEA
- Non-proliferation and Nuclear Security
- Presentation on Safeguards-by-Design and PR
- Presentation on Instrument for Stability
- Export control

Background and Present Status of ESARDA ⇨ Baseline

The RG 2010 reviewed the followig point

History, foundation of ESARDA

Objectives of ESARDA

Current status of ESARDA

- Management structure
- R & D activities of the working groups
- Internal/External communication activities
- Status of implementation of proposals of the Reflection Group 2000

Objectives of ESARDA

At a time ESARDA could broaden the scope of its areas of interest it worth to remind the objectives of ESARDA :

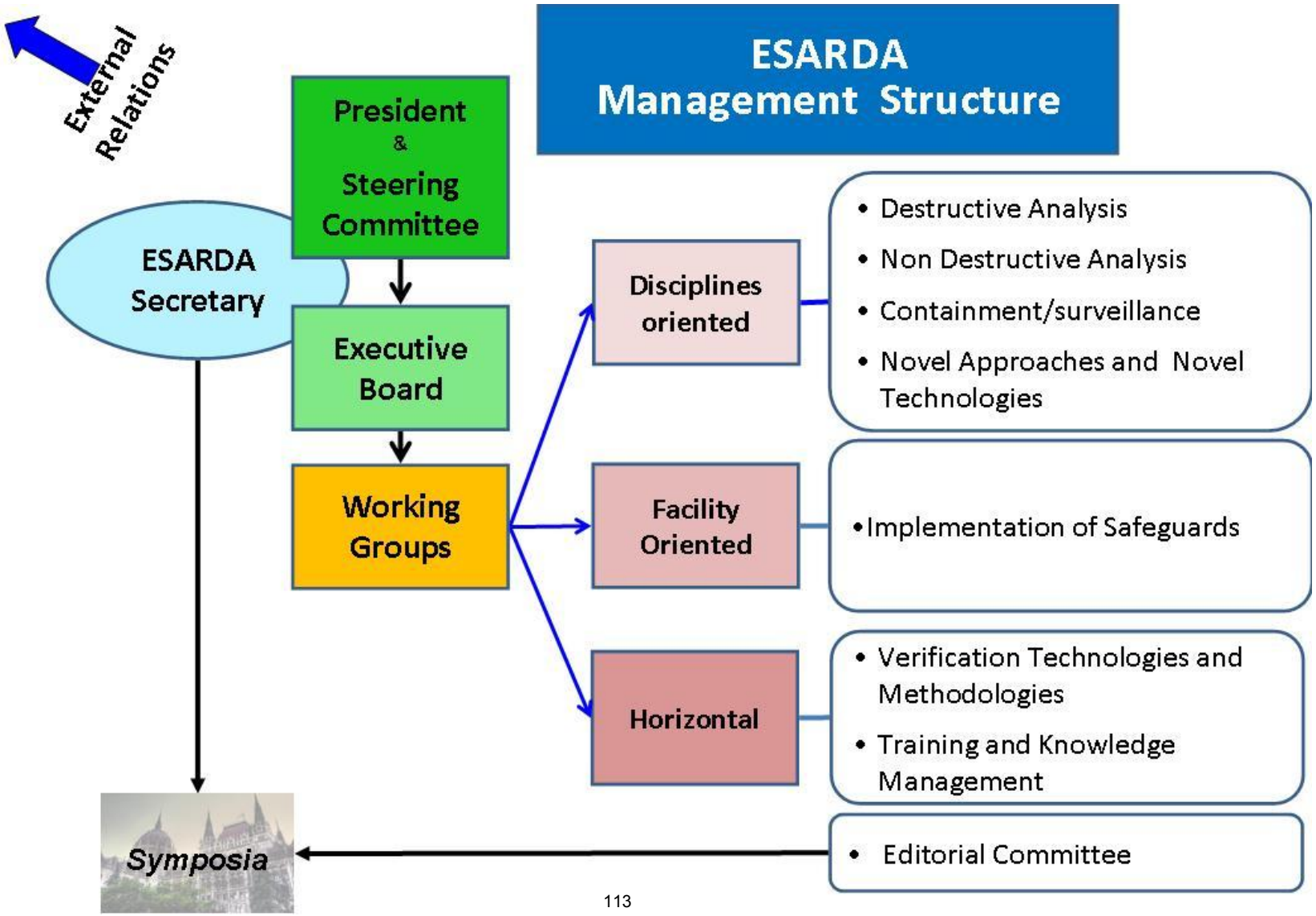
> ***« to advance and harmonise research and development in the area of nuclear safeguards », within international safeguards and to ensure mutual exchange of information and technical assistance to promote the advancement of safeguards, enhancing the efficiency of systems and measures, to investigate on how new techniques can be developed and implemented, to have an educational role and to reach the general public, in particular, those interested in safeguards"***

The RG 2010 considers that the 1969 vision is still valid

ESARDA is a forum for the exchange of information and ideas between nuclear facility operators, safeguards authorities and persons engaged in research and development.

**24 Parties from 15 EU member states and the European Commission,
2 associated members and 7 individual members.**

Management structure



R & D activities of the working groups

The heart of ESARDA lies in its eight working groups. They are recognized as of high technical and scientific value

The RG 2010 assessed the activities of the working groups

Some topics are addressed by several working groups with their specific angle of expertise. This trends will continue in particular with the broadening of ESARDA's scope of its activities.

In that spirit, the common week organized during the fall in Ispra contribute to this coordination

The RG 2010 recommend that a coordination of the activities of the working groups should be done at the management level (Secretariat, Steering) to benefit from the synergy between the WG

Internal/External communication activities

ESARDA's communication is taken care of by the Editorial Committee, responsible for issuing the Bulletin, the symposia's programme and the web-site www.esarda.eu,=.

The RG 2010 found the internal/external communication of ESARDA (e.g. symposia, and topical meetings, ESARDA bulletin, and Web site) fully satisfactory and encourages the Editorial board to pursue a continuous improvements of the quality of these important means to encourage exchange of information and collaboration between members of ESARDA and with the safeguards and non proliferation community at large.

2. Identification of needs and Challenges challenges

Establishment of a new reflection group RG 2010

- From RG 2000 to RG 2010
- Incentive for a new reflection group

ESARDA's forthcoming challenges

- International context in the field of non proliferation and security
- European context in the field of non proliferation and security
- 2010 was a pivotal year for nuclear non proliferation, disarmament and security .
- Nuclear fuel cycle and future development....

Coping to Identified Challenges: ⇨ Response

New framework: a broader role for ESARDA. RG 2010 discussed the following topics

- **Non Proliferation and safeguards**
- **ESARDA and new European policies**
- **Non proliferation vs. nuclear security**
- **Disarmament, fissile material cut-off treaty verification**
- **The evolving nuclear fuel cycle: ESARDA's contribution**

Reshaping ESARDA's R&D activities

Meeting ESARDA's members needs

Coping to Identified Challenges

New framework: a broader role for ESARDA

- ESARDA should continue to be in line with the actual and future international and European security context and address the new verification and safeguards problematic, the extension of the scope of ESARDA deems necessary;

- Nevertheless, the group emphasize the extension of the scope of ESARDA should be done carefully, in agreement with Members States and in the respect of their national responsibilities and confidentiality concerns and with the European Commission;

Current & future topics

ADVANCED SAFEGUARDS — *undeclared materials and activity*

NUCLEAR FUEL CYCLE — *Safeguards-by-Design*

Generation IV Forum

NUCLEAR SECURITY

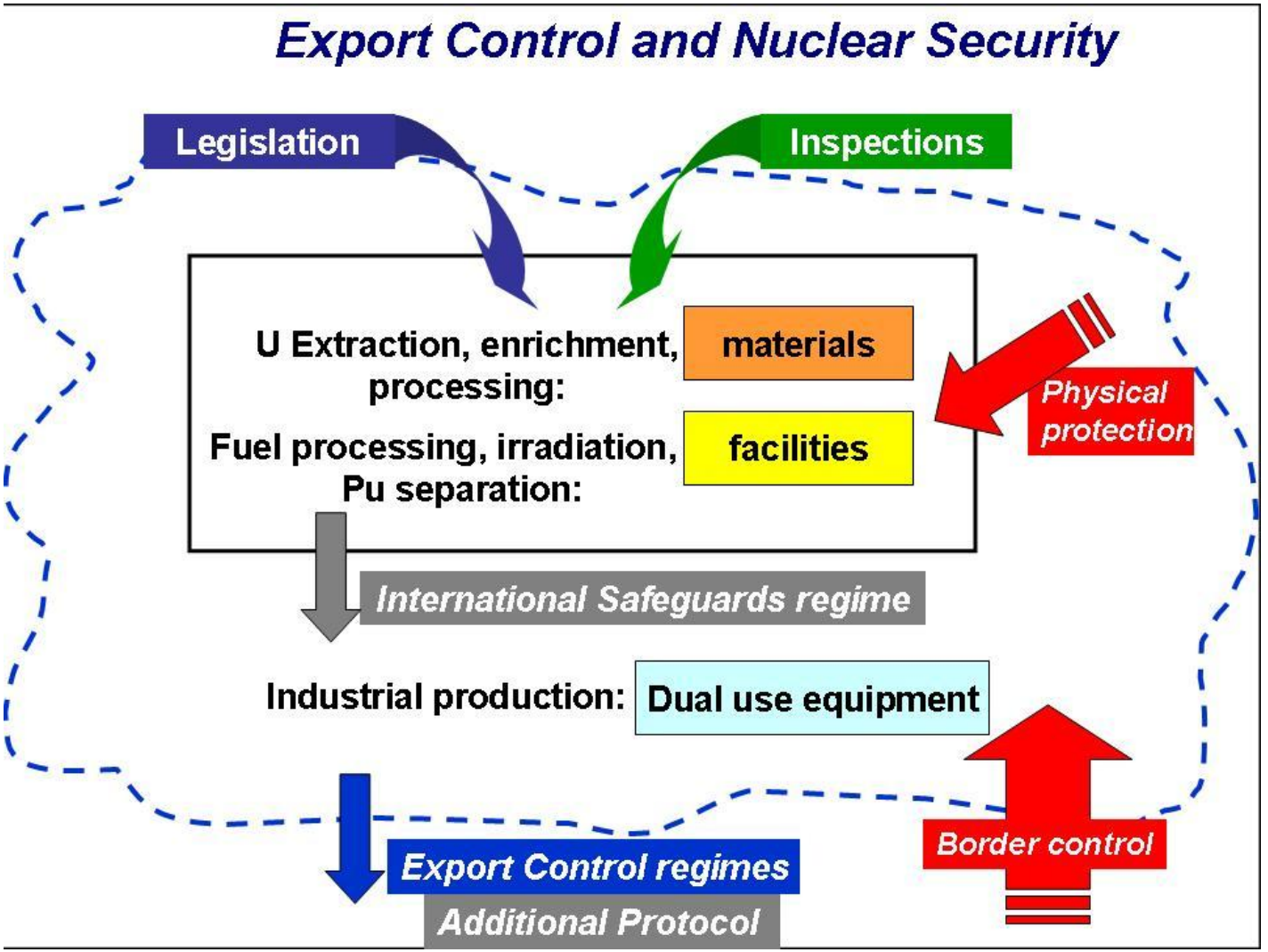
EXPORT CONTROL — *Nuclear Security & Export control*

ILLICIT TRAFFICKING and NUCLEAR FORENSICS

DISARMAMENT: FMCT, CTBT — *Verification*

CYBER SECURITY — *New threats*

EXPORT CONTROL



EURATOM's perspective on the work of ESARDA

In the course of its work the RG 2010 has benefited from the Commission's views on ESARDA

For the Commission ESARDA should continues its work in the framework of the existing Working Groups like WGs on DA, NDA, C/S, IS, TKM, etc., The Commission continue to actively participates in and gives input to these WGs.

ESARDA contributes to the harmonisation of European R&D.

4. Improvement of objectives, scope and management of ESARDA

- Membership and collaborations
- Management
- Continuous reflexion

4. Improvement of objectives, scope and management of ESARDA

Membership and collaborations

- ESARDA should expand its membership base in order to increase its role in the promotion of nuclear security and non-proliferation in the world. in the new Members-States of the Union should also be approached. Academic institutions and Think tanks should also be officially invited to join ESARDA

RG2010 recommends that the Steering Committee evaluate the opportunity and benefits to formalise a cooperation agreement with the INMM. Possible fields of collaboration include the identification of R&D topics for improved Safeguards and Nuclear Security, information exchange, common actions (eg, training, etc)

4. Improvement of objectives, scope and management of ESARDA

Management

RG2010 considers that the present management structure and ESARDA contract serve very well the needs of the association

4. Improvement of objectives, scope and management of ESARDA

Continuous reflexion

The proliferation challenges and scenarios evolve rapidly!

Therefore RG2010 recommends that a reflection process should be held more frequently than present, e.g. every 2-3 years

Conclusion

ESARDA should extend the scope to address new areas in non proliferation and security as expressed previously to allow ESARDA parties, the Commission and the EU to benefit of the recognised unique expertise of its working groups and to stay in line with thr european and european context.

ESARDA has in the working groups all the necessary expertise to address the new challenges.

This extension of the scope of ESARDA should be done carefully in agreement with Members States and in the respect of their national responsibilities and confidentiality concerns and with the Commission:

- ⇨ **Define ESARDA lines of actions/objectives**
- ⇨ **Define the possible areas of actions**

Conclusion

The ESARDA working groups have already started to address some issues internally;

Coordination of the activities of the working groups towards common objectives will enhance ESARDA capabilities and image inside the EU and abroad

External relations with the IAEA should be re enforced as ESARDA as already tight links with the Agency directly or through members states and the Commission.

Cooperation with the "sister" organisation INMM should also developed through the participation to each other working group and set up common project for instances on training

02 IAEA Safeguards System

Making the IAEA Safeguards System Fully Information Driven

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Abstract:

The Department of Safeguards of the International Atomic Energy Agency (IAEA) is continually striving to optimize its processes to improve the effectiveness and efficiency of safeguards implementation. In 2010, the Department completed its first ever Long-Term Strategic Plan covering the period 2012-2023. As part of the development efforts, a structured analysis was performed to determine activities that need to be conducted to prepare for future safeguards implementation challenges and responsibilities. One important direction that cut across a number of the strategic issues identified was the need to further evolve the IAEA's safeguards system to become fully information driven in order to optimize the use of IAEA resources for meeting the safeguards objectives. To this end, the IAEA is further developing the State-level concept for the planning, implementation and evaluation of safeguards activities. State-level approaches will be developed and implemented for all States with safeguards agreements in force taking into account all safeguards-relevant information available to the IAEA about a State, in order to apply an optimized combination of safeguards measures for each State. By taking into account a broader range of State-specific factors, safeguards activities in the field and at Headquarters will become increasingly information driven, focused and efficient. Inspection-related activities and the State evaluation process not only need to be linked but merged. This integration requires (a) changing departmental infrastructure, organization, processes and culture to enhance the exchange of information and encourage collaboration in the evaluation of States and the planning of verification activities, and (b) evolving to a flexible, objectives-based safeguards system that will allow the Department to optimize its safeguards activities. The paper will describe the activities being undertaken by the IAEA to make the IAEA safeguards system fully information driven.

Keywords: IAEA, safeguards, information-driven, State-level, objectives-based

1. Introduction

The safeguards system of the International Atomic Energy Agency (IAEA) provides the international community with credible assurances regarding a State's fulfilment of its safeguards obligations. In a State with a comprehensive safeguards agreement and an additional protocol in force, the broader safeguards conclusion that all nuclear material remains in peaceful activities is based on the IAEA finding no indications of the diversion of declared nuclear material and no indications of undeclared nuclear material or activities in the State as a whole. In a State with a comprehensive safeguards agreement alone, the safeguards conclusion only relates to the non-diversion of nuclear material from peaceful activities. The drawing of such safeguards conclusions is supported by the analysis and evaluation of all information available to the IAEA about a State. In the past two decades, the IAEA's information on States has increased because of additional legal authority (e.g., additional protocols and amended small quantities protocols), improved verification technologies (e.g., environmental sampling and remote transmission of data from IAEA equipment), and enhanced information collection and analysis techniques (e.g., open-source information, satellite imagery and nuclear trade data).

The Department of Safeguards of the IAEA is continually striving to optimize its processes to improve the effectiveness and efficiency of safeguards implementation. In 2010, the Department completed its first ever Long-Term Strategic Plan covering the period 2012-2023. One important direction that cut across a number of the strategic issues identified was the need to further evolve the IAEA's

safeguards system to become fully information driven in order to optimize the use of IAEA resources for meeting the safeguards objectives. In accordance with this strategic plan, the IAEA developed and is implementing an 'action plan' for making the IAEA safeguards system fully information driven. Through this action plan the IAEA is further developing the State-level concept for the planning, implementation and evaluation of its safeguards activities.

Making the IAEA's safeguards system fully information driven requires integrating the IAEA's enhanced State evaluation process, which includes the ability to collect, analyse, and evaluate all information available on a State, with the IAEA's inspection-related activities. At present, information from the inspection-related activities in a State is an important input to the State evaluation; and the IAEA's conclusions, based on the State evaluation, are used in determining the inspection effort for a State. The experience gained over the past decade has shown that for the safeguards system to become fully information driven will require (a) full integration of the State evaluation process with the inspection-related activities and (b) making the safeguards system more objectives based. These constitute the two phases of the action plan.

The core of the IAEA's safeguards system is the State-level concept – a holistic approach to safeguards implementation that is applicable to all States. The State-level concept is based on a comprehensive State evaluation and a State-level approach (which includes a specific combination of safeguards measures for an individual State) that is implemented through a State-specific safeguards implementation plan. Considering the State as a whole provides the opportunity to take State-specific factors into consideration in all stages of safeguards implementation. The safeguards system will be 'fully information driven' when all knowledge on a State is used to focus the appropriate verification effort where needed to achieve the safeguards objectives and to draw safeguards conclusions in the most efficient way.

Knowledge on a State results from the State evaluation process, which is a dynamic, iterative process in which evaluation results constitute the basis for planning safeguards activities, assessing their results, and identifying follow-up actions (e.g., additional information collection or verification activities) required for drawing soundly based safeguards conclusions. Implementation of the State-level concept makes the safeguards system responsive to changes in the analysis, thereby ensuring that the assurances provided to the international community remain credible and up-to-date.

2. Evolving the IAEA Safeguards System

The IAEA safeguards system continues to evolve through enhancing safeguards concepts, processes, approaches, and guidelines that govern the design, implementation, and evaluation of its implementation. State-level approaches, which are currently developed only for States with the broader safeguards conclusion, will be developed and implemented for all States with safeguards agreements in force taking into account all safeguards-relevant information available to the IAEA about a State, in order to apply an optimized combination of safeguards measures for each State. By taking into account a broader range of State-specific factors, safeguards activities in the field and at Headquarters will become increasingly information-driven, focused and efficient. The principles of the safeguards system that is fully information driven, the three levels of objectives that State-level approaches are designed to meet, and the action plan for making the safeguards system fully information driven are described below.

2.1 Principles of the IAEA Safeguards System

The following principles of the IAEA's safeguards system that is fully information driven reflect the integration of the IAEA's safeguards activities and the use of all information known about States.

Differentiation of Safeguards Implementation without Discrimination. The safeguards system will be implemented in a non-discriminatory manner for all States while providing flexibility in determining the optimal combination of safeguards measures for a specific State that comprise the State-level approach. The same generic State-level safeguards objectives will apply to all States that have the same type of safeguards agreement in force. In addition, the same decision-making and evaluation processes will be applied to all States.

Effectiveness as a Priority. The credibility of the assurances provided by the IAEA's safeguards conclusions derives from the effectiveness of the safeguards activities conducted by the IAEA at headquarters and in the field. Safeguards effectiveness will remain a priority that must not at any time be compromised for the sake of seeking cost reductions.

Verification Activities in the Field and at Headquarters. The IAEA's Statute, safeguards agreements, protocols to the agreements, and subsidiary arrangements establish the legal rights and obligations of the IAEA. The IAEA will utilize and make full use of all its legal authority to verify that a State is honouring its safeguards obligations. While information collection and evaluation will be required to achieve objectives related to the State as a whole, nuclear material accountancy will remain of fundamental importance for deriving a conclusion on non-diversion of nuclear material.

Coverage of Acquisition Paths. All plausible acquisition paths for a State to acquire or produce nuclear material for use in a nuclear explosive device are to be covered. Through use of all information known about a State, the plausible acquisition paths for each State will be determined and prioritized. Greater verification effort will be focused on the more critical paths.

Comprehensive Information Analysis and State Evaluation. All safeguards-relevant information available to the IAEA will be comprehensively reviewed, checked for credibility and reliability, and evaluated to enhance the IAEA's knowledge on a State. The information on and knowledge about a State will be used in evaluating the State, preparing the State-level approach, and planning and implementing safeguards activities.

Confidentiality of Information. Departmental information security procedures must protect information while assuring that it is accessible to all staff involved in analysis and evaluation of information on a State. Maintaining the effectiveness and credibility of the Department requires the protection of information on States and related IAEA evaluations, as provided for in safeguards agreements and protocols.

Transparency of Processes. The concept, structure and processes that make the safeguards system fully information driven must be clear to the staff of the Department and to Member States. Member States' confidence in the IAEA's safeguards conclusions will be sustained and enhanced by the Department's transparency on these processes, particularly the processes used for the evaluation of States and for the development of State-level approaches.

Quality Management. Quality management principles will be applied at every stage of planning, implementing. And evaluating safeguards activities. The accuracy of information used in the analysis and evaluation processes is assured through quality controls and audits performed on the safeguards verification, information collection, and information analysis processes.

2.2 Safeguards Objectives

Objectives-based safeguards are based on the following three levels of safeguards objectives:

1. S*afeguards agreement objective.* The objective of safeguards under a comprehensive safeguards agreement is "the timely detection of diversion of significant quantities of nuclear material from peaceful nuclear activities to the manufacture of nuclear weapons or of other nuclear explosive devices or for purposes unknown, and deterrence of such diversion by the risk of early detection." (INFCIRC/153, corrected, Paragraph 28). Each comprehensive safeguards agreement also provides for "the [IAEA's] right and obligation to ensure that safeguards will be applied, in accordance with the terms of the Agreement, on all source or special fissionable material in all peaceful nuclear activities within the territory of the State, under its jurisdiction or carried out under its control anywhere, for the exclusive purpose of verifying that such material is not diverted to nuclear weapons or other nuclear explosive devices." (INFCIRC/153, corrected, Paragraph 2). The objectives for States under item-specific safeguards agreements (INFCIRC/66) and voluntary offer agreements differ from that of a comprehensive safeguards agreement.

2. *Generic State-level safeguards objectives.* The generic State-level safeguards objectives for States with comprehensive safeguards agreements are: (a) to detect undeclared nuclear material and activities in the State as a whole; (b) to detect undeclared production or processing of nuclear

material in declared facilities and locations outside facilities (LOFs); and (c) to detect diversion of declared nuclear material in declared facilities and LOFs. The first objective is not applicable to States with a voluntary offer agreement or to States which are not party to the Nuclear Non-Proliferation Treaty. It should be noted that the IAEA has insufficient tools to fully achieve the first objective for a State with a comprehensive safeguards agreement that does not have an additional protocol in force.

3. *State-specific technical objectives*. State-specific technical objectives are the third-level of safeguards objectives and must be attained to accomplish the relevant generic State-level safeguards objectives. The technical objectives are associated with the plausible acquisition paths for the proliferation of nuclear explosive devices as determined from State-specific factors and an acquisition path analysis. The technical objectives form the basis for determining the State-level safeguards approach.

2.3 Action Plan

Making the IAEA's safeguards system fully information driven will be implemented through the following two phases which are described in subsequent sections.

1. Changing the departmental infrastructure, organization, processes and culture to enhance the exchange of information and to encourage collaboration in the evaluation of States and the planning of verification activities. Phase 1 is to be accomplished during 2011, the first year of the action plan.

2. Enhancing the safeguards processes to produce a safeguards system that is more flexible and objectives-based and will allow the Department to optimize better its safeguards activities. Phase 2 is to be accomplished by the end of 2012, the second year of the action plan.

3. Infrastructure, Organization, Processes, and Culture

In order to maintain effectiveness, the Department's infrastructure, organization, processes, and culture must support collaborative work with enhanced intra-departmental and, as appropriate, inter-departmental exchange of information to establish the most complete and accurate knowledge about States. Departmental management, at all levels, plays a critical role for ensuring the effectiveness of the safeguards system. Management must encourage collaborative work and proactive involvement of staff to achieve continual improvement of the Department's operations and capabilities in order to effectively carry out the IAEA's verification mission.

3.1 Information Management

For the safeguards system to be fully information driven, the information management system must contain those structures necessary to make all information on a State, including results from State evaluations, in-field activities and transnational issue analysis, available to staff performing verification, analysis and evaluation activities for the State. This integrated information management system will include analytical capabilities and geospatial data representations, among other information technologies that will provide all staff involved in safeguards implementation and collaborative analysis with access to all relevant information and knowledge available on the State. This infrastructure will facilitate the cross-divisional exchange of information, expertise, experience, and best practices, as well as make available tools for data and information analysis.

3.2 Documenting and Reporting Verification Activities

The primary State-specific documents relevant to a State are the State evaluation report, the State-level approach, and the State-specific implementation plan. The results of the analyses performed in the preparation of each document affect the information contained in the other documents. Thus, the process for preparing these documents will be integrated to ensure persons involved in their development and preparation are informed with respect to all State information, to promote

consistency between the documents, and to minimize the effort required to prepare and maintain the documents.

3.3 Staffing and Culture

The Department's personnel are a major component of the safeguards system's effectiveness. The Department of Safeguards must be a flexible and responsive organization that learns from its experiences, is knowledge-based, and provides incentives for enhancement of staff capabilities. Recruitment practices will be reviewed and strengthened to ensure that the Department acquires staff with the necessary qualifications that will make them able to contribute to the effectiveness of the safeguards system. In addition, the Department will support continual staff development by implementing an advanced training programme that reflects career path development and addresses the needs of staff members, in particular country officers, senior inspectors and information analysts. Training programmes will be updated to establish a common departmental understanding of the objectives-based process and for implementing collaborative processes.

4. Enhancing the Processes of the Safeguards System

The processes of the safeguards system involve the use of knowledge about States to determine State-level approaches, which in turn are used to determine the safeguards planning and implementation activities necessary to draw safeguards conclusions, which are based on evaluation of all information available to the IAEA. Figure 1 illustrates the integration of the safeguards system's processes which are described in the following sub-sections.

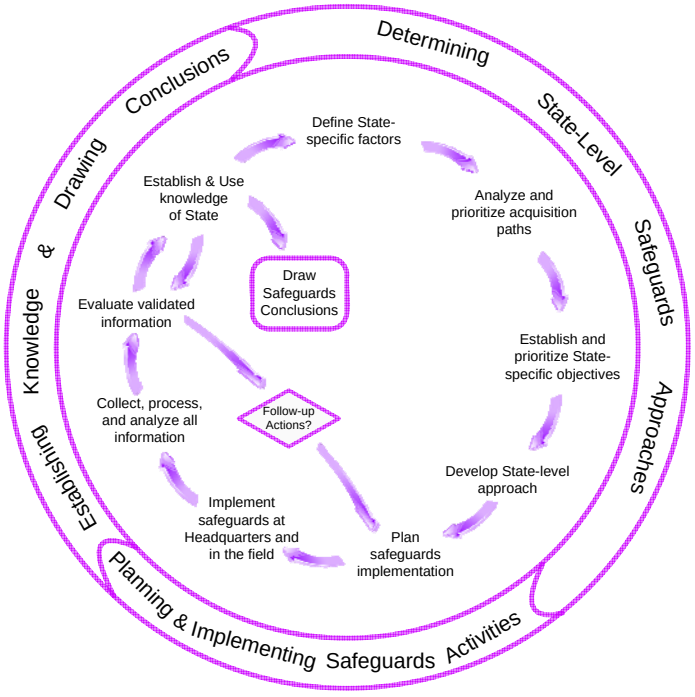

Figure 1. Integration of processes of a safeguards system that is fully information driven

4.1 Processes Establishing Knowledge of a State and Drawing Safeguards Conclusions

Knowledge about a State is developed through the structured accumulation and use of all evaluated information about a State, including the State's association with transnational activities and potential involvement in illicit trade. Acquisition of this knowledge is to be enhanced through collaborative processes that involve all organizational units of the Department of Safeguards. Because this is an iterative and ongoing process, the knowledge about a State will continually expand and improve.

Collect, process, and analyse all information. The Department collects all information on States (including transnational interactions) on an ongoing basis. This information is obtained through (a) reports and declarations made by the State; (b) the results of IAEA in-field and Headquarters verification activities (including inspections, complementary access, environmental and destructive analysis sample reviews, material balance evaluations, and remote monitoring reviews, among others); (c) information collected from open sources (including satellite imagery); and (d) information provided by other sources such as persons, companies, organizations, and States (including nuclear-related trade information). Information from all sources will be continually processed and analysed. This analysis of information includes assessments of inspection results, sample data, and the credibility and reliability of collected information.

Evaluate Validated Information. The evaluation of a specific State is performed by a Core State Evaluation Group (CSEG) using collaborative processes to establish the IAEA's knowledge about the State. CSEGs ensure a comprehensive evaluation of all information available; plan and report on an ad hoc basis, including the preparation of State Evaluation Reports (SERs); determine State-specific factors; and provide input to the Divisions of Operations for preparing State-level approaches, when applicable.

Use of knowledge about a State. Knowledge, which is developed through the evaluation of all valid and analysed information available to the IAEA on a State, is used to (a) evaluate further information obtained by the IAEA, (b) draw safeguards conclusions, and (c) develop State-level approaches.

Draw Safeguards Conclusions. The State evaluation reports, which contain recommendations on safeguards conclusions, are reviewed periodically by the Department. The knowledge that is developed through the State evaluation process forms the basis for the safeguards conclusions on a State's compliance with its safeguards obligations. The safeguards conclusion for a State is made by a committee of senior departmental managers.

4.2 Processes Determining State-Level Approaches

The continuously increasing knowledge about a State, combined with the safeguards conclusions for the State, is to be more effectively used to determine the safeguards activities that need to be performed to close gaps in knowledge, or to enhance or maintain knowledge about the State. State-specific factors in combination with the plausible acquisition paths identified for a State are used to define and prioritize State-specific technical objectives. The optimal set of safeguards measures to accomplish the State-specific technical objectives for the State comprises the State-level approach.

Define State-specific factors. State-specific factors are a compilation of all the characteristics of a State that are relevant to the development, implementation, and evaluation of safeguards for a State. Certain State-specific factors, such as the nuclear fuel cycle capabilities of the State, are to be used in the acquisition path analysis. Others, such as multinational ownership of facilities in the State, are to be used to determine the priority of the plausible acquisition paths. Others would influence the State-specific technical objectives, the priority of the technical objectives, the performance targets, or the selection of safeguards measures. Other State-specific factors, such as other non-proliferation undertakings, may be considered in the State evaluation process.

Analyse and prioritize acquisition paths. For the IAEA to detect efficiently the potential diversion of declared nuclear material, misuse of nuclear facilities, or presence of undeclared installations supporting the manufacture of nuclear explosive devices, the IAEA must determine the plausible paths by which a State might acquire the nuclear material for such devices. A State's nuclear fuel cycle capabilities should be assessed in terms of the State's (a) existing nuclear fuel cycle, (b) knowledge of and expertise on nuclear fuel cycle technologies (including through past and current research and development activities), (c) experience with operating related processes and facilities (e.g., enrichment of non-nuclear isotopes), (d) capacity to manufacture or import the technology needed, (e) ability to produce or import the feed materials for the processes, and (e) time and resources to develop necessary capabilities. The assessment of a State's capability to operate undeclared facilities and the ability to acquire feed materials for such facilities contribute to the plausibility of the potential acquisition paths. Some acquisition paths will be judged to be more likely than others; however, all plausible acquisition paths are to be considered and covered.

Establish and prioritize State-specific technical objectives. Each potential acquisition path has multiple points at which its use could be detected. Because multiple acquisition paths may pass through common processes, evaluating the set of plausible paths will reveal those processes common to several paths and those points on the paths that are most critical for detecting the use of the paths. State-specific technical objectives are to be established for each plausible acquisition path to identify what must be detected with respect to diversion of declared nuclear material, detection of misuse of facilities, or detection of other undeclared activities.

The technical objectives are to be prioritized using State-specific factors and the relative importance of the objective on the acquisition paths for the State. The potential State-specific technical objectives will be evaluated to determine the optimal combination of objectives that would provide for the most efficient use of IAEA resources in providing coverage of the plausible acquisition paths.

For each State-specific technical objective, performance targets will be established that ensure that the planned safeguards measures achieve the generic safeguards objectives. The performance targets for the diversion-relate objectives may include State-specific timeliness goals, quantity goals, and detection probabilities.

Develop State-level approach. To achieve each of the State-specific technical objectives, safeguards measures will be identified that could achieve the technical objective at the level specified by the performance targets. These measures include inspections, design information verification visits, complementary access, sample taking and analysis, information collection and analysis, satellite imagery analysis, use of advanced verification technologies and techniques, and the use of unattended monitoring systems and remotely monitored data, among others. The selection of measures will take into consideration State-specific factors such as the capabilities of the State or regional safeguards authorities. The potential State-specific combinations of technical measures will be evaluated taking into consideration the State-specific factors and the cost of each alternative approach to determine which combination has the best cost-benefit value. The optimal combination of safeguards measures becomes the State-level approach.

4.3 Processes Planning and Implementing Safeguards in the Field and at Headquarters

Headquarters and in-field activities are defined by a State-specific implementation plan that is derived from the State-level approach.

Plan safeguards implementation. A State-specific implementation plan that defines the activities needed to meet the performance targets of the State-level approach will be developed for each State with a safeguards agreement in force. The selection of the activities will take into consideration State-specific factors and results of the State evaluation. The State-specific implementation plan is to be revised as necessary to respond to new information received and findings from State evaluations. Activities specified in the plan will be revised, when and as necessary, to address not only anomalies and discrepancies but also changes in State-specific factors identified through the continuous State evaluation process.

Implement safeguards at Headquarters and in the field. The safeguards activities are conducted by qualified inspectors and analysts both in the field and at Headquarters with sharing of information and knowledge. Departmental policies, guidance and procedures will continually be updated to describe the requirements and best practices for how the safeguards implementation activities should be conducted. In implementing safeguards in the field and at Headquarters, effectiveness and efficiency gains will continue to be sought from the use of technological and scientific advancements in safeguards techniques and equipment. When a planned safeguards measure cannot be performed in accordance with the State-specific implementation plan, an evaluation will be made to determine if alternative information or measures are available for meeting the State-specific technical objective, or if additional activities must be planned.

5. Conclusion

Making the IAEA's safeguards system fully information driven requires (a) changing departmental infrastructure, organization, processes and culture to enhance the exchange of information and collaboration in the evaluation of States and the planning of verification activities, and (b) evolving to a flexible, objectives-based safeguards system that will allow the Department to optimize its safeguards activities. The Department of Safeguards is implementing an action plan to be implemented over the next two years. A strategy is being developed for communicating the evolution of the safeguards system to staff and to Member States to ensure that the changes are understood, adopted and sustained. Transparency on the transition to objectives-based safeguards will be provided through reports to Member States, such as the annual report, the safeguards implementation report, the report on strengthening the IAEA safeguards system, and via the IAEA website.

The IAEA Safeguards System – Providing Credible Assurances

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Abstract:

One of the International Atomic Energy Agency (IAEA)'s key objectives is to deter the proliferation of nuclear weapons by detecting early the misuse of nuclear material or technology and by providing credible assurances that States are honouring their safeguards obligations. This paper describes the processes through which the international community can be assured that the IAEA's safeguards conclusions are soundly based – these include: a rigorous and ongoing evaluation of all information available to the Agency about States' nuclear programmes and other activities of safeguards relevance by collaborative, multidisciplinary teams of Agency experts; detailed reporting to, and review by, internal review committees; independent annual review of selected State Evaluation Reports; and thorough, ongoing evaluation of the effectiveness of safeguards verification activities in the field and at headquarters. All of these processes are based on sound, well-documented procedures and are subject to internal quality audit. The paper also discusses recent initiatives within the IAEA's Department of Safeguards to strengthen these processes.

Keywords: safeguards; safeguards conclusions; State evaluation process; State-level concept; safeguards evaluation

1. Introduction

One of the International Atomic Energy Agency (IAEA)'s key objectives is to deter the proliferation of nuclear weapons by detecting early the misuse of nuclear material or technology and by providing credible assurances that States are honouring their safeguards obligations. To meet this objective effectively and efficiently, the IAEA's safeguards system must continue to evolve. In 2010, the Department of Safeguards completed its first ever long-term strategic plan and is in the process of developing and implementing strategies to optimize its processes and the way it works. These include developing the conceptual framework, fully exercising the IAEA's legal authority, enhancing the IAEA's technical capabilities, strengthening cooperation and collaboration, and managing its human and financial resources.

The conceptual framework for the IAEA safeguards system has been evolving since the implementation of the State-level approach in the early 2000s in States for which a broader conclusion had been drawn. A State-level approach defines the State-specific technical objectives needed to meet the three generic State-level objectives.[1] It is based on a thorough acquisition path analysis to determine the most likely pathways a State could use to acquire weapons-usable nuclear material, and tailors the safeguards activities at Headquarters and in the field in ways necessary in order to detect such material and activities. The State evaluation process seeks to answer the question

[1] Objective A: to detect undeclared nuclear material and activities in the State as a whole; Objective B: to detect the undeclared production or processing of nuclear material in declared facilities and locations outside facilities (LOFs); Objective C: to detect the diversion of declared nuclear material in declared facilities and LOFs.

whether all relevant information about a State's nuclear programme is complete and consistent. It thus is an important component of the conceptual framework and the main process used by the Department to derive safeguards conclusions.

2. Safeguards agreement objectives and safeguards conclusions

Article III of the Treaty on the Non-Proliferation of Nuclear Weapons (NPT) requires each non-nuclear weapon State party to the Treaty to conclude a comprehensive safeguards agreement (CSA) with the IAEA, under which safeguards are applied on all nuclear material in all peaceful nuclear activities within the territory of the State, under its jurisdiction, or carried out under its control anywhere. [1]

The objective of safeguards under a CSA is "the timely detection of diversion of significant quantities of nuclear material from peaceful nuclear activities to the manufacture of nuclear weapons or of other nuclear explosive devices or for purposes unknown, and deterrence of such diversion by the risk of early detection." [2] Each CSA also provides for "the Agency's right and obligation to ensure that safeguards will be applied, in accordance with the terms of the Agreement, on all source or special fissionable material in all peaceful nuclear activities within the territory of the State, under its jurisdiction or carried out under its control anywhere, for the exclusive purpose of verifying that such material is not diverted to nuclear weapons or other nuclear explosive devices." [3]

In 1997, the Board of Governors approved the text of the Model Additional Protocol [4] to "strengthen the effectiveness and improve the efficiency of the safeguards system as a contribution to global nuclear non-proliferation objectives". When adopted by a State, an additional protocol (AP) expands the IAEA's access to information and locations in the State and thereby significantly strengthens the IAEA's ability to verify the peaceful use of all nuclear material in that State.

The safeguards conclusion the IAEA is able to draw for a State depends on the type of safeguards agreement in force. For a State with a CSA alone, the IAEA strives to conclude that there is no indication of diversion of declared nuclear material from peaceful nuclear activities in the State. Besides conducting a comprehensive evaluation of all safeguards-relevant information available to the IAEA, the focus of verification activities is mainly on the correctness of declarations on nuclear material at declared facilities. While the IAEA has the right and obligation under a CSA to verify not only the correctness but also the completeness of State declarations, its tools to do so are limited without an AP in force.

For a State with both a CSA and an AP in force, the IAEA has more information and more tools at its disposal to conclude that there is no indication of the diversion of declared nuclear material and of undeclared nuclear material and activities in a State. Before doing so, the IAEA will conduct a comprehensive State evaluation based on all information available to the Agency about the State's nuclear and nuclear-related activities (including declarations submitted under the AP); implement complementary access, as necessary, in accordance with the State's AP; and address all anomalies, questions and inconsistencies identified in the course of its evaluation and verification activities. If the IAEA has found no indication of the diversion of declared nuclear material from peaceful activities, and no indication of undeclared nuclear material and activities, it can draw the so-called broader conclusion for that State, i.e. that that all nuclear material remained in peaceful activities. The broader conclusion is the pre-condition for the implementation of integrated safeguards, which optimizes safeguards activities at Headquarters and in the field leading to gains in effectiveness and efficiency.

Drawing conclusions regarding the absence of undeclared nuclear material and activities is intrinsically more challenging than verifying the absence of diversion of declared nuclear material. The expectation of Member States that this conclusion be soundly based, and thus provide credible assurance that a State is meeting its safeguards obligations, requires the Agency to study very carefully how the conclusion is derived.

3. Rigorous and ongoing State evaluations

Starting from the mid-1990s, IAEA safeguards have evolved from facility-based verification and evaluation activities to a system that uses all information available to the IAEA on a State as the basis for drawing safeguards conclusions. The State evaluation process has become the heart of this system. The main purpose of State evaluations is to assess the consistency of States' nuclear programmes with all other information available to the IAEA and to identify early any indications of undeclared nuclear activities or material. This consistency analysis is carried out by evaluating information declared by the State (for example, nuclear material accounting reports, design information, AP declarations or any other information provided by the State) against all other information available to the IAEA. This information comprises records from in-field activities (e.g. reports on inspections, complementary access and design information verification visits, containment and surveillance data, analytical results of samples taken at nuclear facilities), IAEA internal information, mainly originating from IAEA databases, information from a variety of open sources (scientific and technical literature, news reports, satellite imagery), and any information provided to the IAEA by third parties. Any inconsistencies identified in the course of the State evaluation are flagged and followed up by activities at Headquarters and/or in the field.

State evaluations are conducted in an ongoing manner for most States with safeguards agreements in force. This work is carried out in a collaborative manner by intradepartmental State Evaluation Groups (SEGs), consisting of the country officer from the Division of Operations directly responsible for the State under evaluation, and specialized staff from different sections of the Department of Safeguards responsible for analysing the information received and/or collected by the IAEA (for example, open source analysts, satellite imagery analysts, nuclear material and environmental sample analysts). This ensures that all expertise available within the Department is fully utilized and integrated into the analytical process.

The results of State evaluations are periodically documented in State evaluation reports, which are reviewed and assessed by two interdepartmental review committees, the Information Review Committee (IRC) and the Information Review Sub-committee (IRS-C). These reviews form the basis of safeguards conclusions, which are published in the annual Safeguards Implementation Report (SIR).

The IRC reviews a small number of States per year and is composed of directors from within the Department of Safeguards and the Director General's Office. These are either States with ongoing safeguards issues or States for which the broader safeguards conclusion is being drawn for the first time. The Agency draws such a conclusion when a State has both a CSA and an AP in force and after a thorough information review has shown no indication of diversion of declared nuclear material or of undeclared nuclear material or activities in the State.

The membership of the IRS-C consists of section heads, senior inspectors and senior analysts. The IRS-C convenes two to three times per month and reviews the majority of State evaluation reports (currently about 120 per year). State evaluation reports are prepared yearly for States with an AP in force and for most States of proliferation concern. For States without an AP in force, in particular for the large number of States with limited nuclear activities and with a small quantities protocol [5] in force, State evaluation reports are prepared on a less regular basis. No matter whether States are formally evaluated by the information review committees or not, State evaluations are conducted by the State evaluation groups (SEGs) on an ongoing basis, to ensure the timely detection of inconsistencies and proliferation indicators and the need for follow-up actions.

4. Recent initiatives to enhance the State evaluation process

Due to its importance for the work of the Department of Safeguards, the State evaluation process has been reviewed twice in recent years in order to identify ways and means for its improvements: in 2007, a working group established by the Department of Safeguards' Deputy Director General submitted several suggestions for improvement of the process. In 2009, the IAEA's Office of Internal Oversight (OIOS) carried out an assessment of the State evaluation process and came up with further recommendations. In 2010, several actions, as recommended by the two groups, were implemented to enhance the State evaluation process. They comprise: the introduction of a prioritization scheme for

State evaluations; the empowerment of SEGs; the creation of a 'Red Team' to assure the quality and sound basis of safeguards conclusions; and the establishment of a roster of experts within the Department of Safeguards.

In order to better focus the Department's analytical resources, a prioritization scheme for State evaluations was recently introduced. Under this scheme, interim State evaluation reports (ISERs) are produced for certain States, based on factors such as type of safeguards agreement in force, years of successful implementation of integrated safeguards et cetera. The ISERs are concise and issues-driven, focusing on the follow-up actions carried out on previous review committee recommendations and on newly arising matters of safeguards significance. While the ISERs are substantially shorter than standard State evaluation reports, ongoing State evaluation by SEGs is particularly important in order to ensure that all information is evaluated for its safeguards relevance as soon as it becomes available. The prioritization scheme will allow for a more efficient and focused review of the ISERs by the review committees. It will free up the Department's analytical resources so that they can be focused on States with larger nuclear fuel cycles or safeguards issues.

Another initiative was the 'empowerment' of 12 SEGs responsible for States with larger nuclear fuel cycles. The SEG members received special training on enhanced collaborative information analysis. They were then given the task to try out new ways of cooperation when evaluating the States under their responsibility, utilizing the individual expertise of the SEG members to the benefit of the evaluation, with the objective of preparing enhanced State evaluation reports for those States. Every SEG was permitted to choose the work methodology that suited them best. Some of the SEGs created a virtual work space on the Safeguards Portal, the internal web site of the Department of Safeguards, which enabled them to share all information on the States across different divisions on a need-to-know basis and to use this tool for the actual drafting of the State evaluation report. It also allowed for communication amongst the SEG members, no matter whether they were at Headquarters or in the field. Other SEGs met physically on a regular basis and exchanged thoughts, ideas and analytical results. The result of this enhanced collaborative analytical effort was 12 State evaluation reports that were distinctly different from other reports. Some of them made use of probabilistic assessments, a methodology that had never before been employed in the Department of Safeguards. Others used graphs to present analytical results of numerical data. The analysis was thorough and comprehensive, utilizing all subject-specific expertise. It is now envisaged that this process of continuous, collaborative analysis will be deployed across the entire Department of Safeguards.

An evaluation of the lessons learned from the 'empowered' SEGs has highlighted the need for actions that will affect the resources, information and processes involved in State evaluation work. With regard to the resources, the process is very labour-intensive and an optimized composition of SEGs and resource management will be vital, as will be the training and the enhancement of analytical skills of all staff involved in State evaluations.

With respect to information, the lack of standardized, user-friendly virtual space arrangements for sharing information across the Department on a need-to-know basis constitutes a major obstacle for performing collaborative analysis efficiently. The high-level requirements for a tool to facilitate such collaboration, the Virtual State File (VSF), have already been defined and the implementation of the VSF is dependent on the completion of the ISIS Re-engineering Project (a project to migrate the IAEA Safeguards Information System (ISIS), which is currently based on out-dated information technology, to a modern environment). Once deployed, the VSF will provide a single point of entry to all State-related information with controlled and secure access. It will simplify teamwork and information sharing across the multiple divisions that contribute to ongoing State evaluations. Until then, intermediate solutions will need to be sought.

As far as the processes are concerned, the current State evaluation process is largely driven by the need for preparing a report and the associated review committees' review schedules. In the future, a methodology will need to be developed to facilitate the preparation of the reports while moving to a continuous process of information collection, evaluation and analysis. A more issues-driven approach would also be desirable.

The 2009 evaluation by OIOS concluded that the final product of each State evaluation, the State evaluation report, was provided to the interdepartmental review committees without any quality assurance measures during the process. OIOS recommended the inclusion of a quality assurance

step in the State evaluation process, to ensure that all information available to the IAEA had been considered for the analysis and that the recommendations proposed in the respective State evaluation reports were indeed soundly-based.

As a consequence, an independent departmental evaluation team, the so-called 'Red Team', was established in May 2010 to review, on a trial basis, already completed State evaluation reports as a quality assurance measure for a small number of reports. The objectives of the Red Team review were to identify gaps in the information collected; to recognize weaknesses in the analysis of the information; to consider the effect of any such gaps or weaknesses on the reliability of the safeguards conclusions drawn; and to identify weaknesses or other issues generic to the State evaluation process.

The Red Team concluded that, while no specific issues were identified for the States reviewed that would call into question the safeguards conclusions drawn, there was room for improvement. Several generic recommendations, identified by the Red Team, are currently being evaluated. The quality assurance concept will be incorporated into the State evaluation process. Further issues to be considered are, for example, the number and selection of the State evaluation reports to be reviewed, membership and work prioritization of the Red Team, and the implementation of the generic recommendations identified in the Red Team report.

Another recommendation resulting from the two review teams' recommendations was the introduction of a roster of experts. This roster consists of departmental staff who possess specific expertise in the areas relevant for State evaluations. About 80 staff members volunteered to put their knowledge and skills at the disposal of SEGs. The outcome of this effort will be monitored and evaluated.

5. Evaluation of the effectiveness of safeguards activities

Within the Department of Safeguards, the effectiveness of safeguards implementation and of verification activities at the facility level and at the State level is evaluated by the Section of Effectiveness Evaluation (SEE). The results of this evaluation are reported in the SIR.

SEE presently evaluates the extent to which safeguards objectives for each facility and State have been attained in a given year. In the future, SEE will also evaluate the effectiveness of the State evaluation process, for example by including a quality assurance step in the process, as initiated in 2010 by the use of the Red Team.

6. Procedures, internal quality audit and quality management system

The Department of Safeguards has undertaken great efforts to map all departmental processes, both at Headquarters and in the field. A large number of staff involved in the processes were interviewed for this purpose. The process maps were found to be very useful for documenting all steps involved in each of the processes and for preparing respective procedures. They are also used for assigning costs to each of the process steps, which is helpful for budgeting purposes as well as for optimizing processes.

Internal quality audit procedures have been developed, and audits are carried out on a regular basis, for selected departmental processes. Since the introduction of the system in 2006 more than 20 departmental processes have been reviewed. For example, the entire process of environmental sample taking was audited, covering the preparation of the sampling kit at the Safeguards Analytical Laboratories (SAL), the actual sample taking and handling by inspectors in the field, the dispatch of the samples from the field to SAL, the processing of samples at SAL, the evaluation and analysis of the sample results and the provision of these results to the respective Division of Operations. The audits ensure the quality of the processes and a uniform adherence to procedures across the Department; the results of the audits are used to further improve the processes.

In light of the changing safeguards environment and the move from quantitative to more qualitative assessments, the need for a comprehensive quality management system was recognised by the Department of Safeguards. A quality management system based on ISO 9001:2000 is now being implemented within the Department, which further increases the confidence in the soundness of safeguards conclusions. [6]

The changes underway at the Department of Safeguards will result in major modifications to established processes and the need to update current procedures and internal quality assurance measures.

7. Conclusion

With the full implementation of the conceptual framework for the IAEA safeguards system and the move to a safeguards system that is fully information-driven, the Department of Safeguards' processes and procedures will undergo a major change. As described in this paper, several measures have already been put in place to facilitate this change. An action plan has been developed to address all of the requirements necessary to effect change. The result of these efforts will be a safeguards system that is objectives-based and that will fully integrate activities at Headquarters and in the field, thus continuing to provide credible assurance to the international community in an effective and efficient manner.

8. References

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[3] *Ibid*., para. 2.

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[5] *Small quantities protocols to comprehensive safeguards agreements are concluded between the IAEA and a State on the basis that the State has less than specified minimal quantities of nuclear material. This protocol holds in abeyance the implementation of most of the detailed provisions of Part II of the CSA until such time as the quantity of nuclear material in the State exceeds the prescribed limits or the State has nuclear material in a facility* as defined in INFCIRC/153.

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The European Commission Cooperative Support Programme: 30 Years of Activities

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Abstract:

The IAEA bases its technical and scientific Programme on contributions from the Member State Support Programmes (MSSP). The European Commission Cooperative Support Programme (EC-SP) started in 1981 to support IAEA's activities in the field of nuclear safeguards. Since its beginning, the EC-SP has been operated by the European Commission's Joint Research Centre (JRC) and its institutes at Ispra-Italy, Geel-Belgium and Karlsruhe-Germany. The EC-SP tasks provide technology and expertise in many technical areas related to the effective implementation of safeguards verification measures including the detection of undeclared materials, activities, and facilities. The paper details the main activities of the EC-SP in recent years in terms of the specific work as part of tasks with well-defined milestones and deadlines, training activities and the technical consultancy support to the many IAEA meetings and expert groups.

Keywords: IAEA, Support Programme, EC-SP

1. Introduction

The European Commission Cooperative Support Programme to the IAEA in the field of research and development in Nuclear Safeguards – EC-SP – was officially created on the 7th of May 1981 with an exchange of letters between Directors of the European Commission and the IAEA. Since then the EC-SP has been involved in more than 115 tasks in different technical and application areas of Nuclear Safeguards. In 2011, the EC-SP celebrates its 30th anniversary.

This paper details the main EC-SP activities in the last 30 years of activities. It starts with some historical background and description of the current modes of operation, including the close collaboration with the European Commission's Directorate General on Energy – ENER, in charge of the implementation of the EURATOM treaty. The paper then highlights some recent achievements of the EC-SP and ends with some discussion on current practices and future.

2. Historical Background

The IAEA was created in 1957, the same year as the Treaty of Rome (instituting the European Economic Community) and the EURATOM Treaty (instituting the European Atomic Energy Community) were signed. As a consequence of the EURATOM Treaty, an executive Commission of EURATOM (later merged into the Commission of the European Communities which later became the current European Commission) was mandated to implement the EURATOM Treaty, including all Nuclear Safeguards and verification measures.

In 1970 the Nuclear Non-Proliferation Treaty – NPT – entered into force and the IAEA received the mandate to create and implement an International Nuclear Safeguards regime.

Considering the technical character of Nuclear Verification methodologies, there was much technical collaboration between the IAEA and the European Commission's Joint Research Centre – which had

been created in 1959 with the specific role of fostering joint European research in nuclear energy related matters.

After the creation in 1977 of the Member States Support Programme – MSSP, the European Commission joined the MSSP on the 7th of May 1981 with an exchange of Letters establishing a "formal Cooperative Support Programme between the IAEA and EURATOM in the field of Research and Development in Safeguards". The signatories were Messrs Sigvard Eklund, Director General of the IAEA, and Wilhelm Haferkamp, the German Commissioner for External Relations including Nuclear Affairs of the Commission of the European Communities (President: Gaston Thorn).

The exchanged letters indicated that "… the programme will cover the following areas of R&D activity":

a) Surveillance and containment

b) Measurement technology

c) Training Courses

d) Information data, treatment and evaluations

3. EC-SP Modes of Operation

The European Commission's Joint Research Centre (JRC) operates the EC-SP. Two JRC institutes with a work programme in the field of Nuclear Safeguards are actively collaborating with the IAEA under the framework of EC-SP. These are:

- Institute for Reference Measurements and Materials (IRMM), Geel, Belgium
- Institute for Transuranium Elements (ITU), Karlsruhe, Germany

The European Commission Directorate General on Energy – ENER, in charge of the implementation of the EURATOM Treaty, is kept informed about all IAEA requests as well as with the progress and implementation of current tasks. On a case by case basis, and whenever appropriate, ENER proposes trilateral collaboration schemes for the execution of specific tasks.

As any other MSSP, IAEA's Support Programme Coordination Group meets twice a year with the coordinator of the EC-SP and specific task officers for overall task review meetings.

3.1 Research and Development Tasks

The different meetings between JRC and IAEA staff contribute to a widespread dissemination of knowledge:

- JRC staff is aware about IAEA needs and orientations.
- IAEA staff learns about recent research activities, including new R&D results, laboratories, equipment, investments, etc.
- The regular MSSP coordinator meetings and IAEA R&D reports also contribute to this exchange of knowledge

These informal bilateral exchanges are beneficial as they contribute to bring together end-users and developers. Further, the good understanding of IAEA needs often influence future JRC multi-annual work programmes. On an annual basis, JRC's internal definition of work-programme objectives and deliverables for the different groups also reflect the current IAEA tasks.

3.2 Expert Meetings and Workshops

JRC staff regularly, often together with colleagues from ENER, participates to meetings, expert networks, workshops, etc. organised by the IAEA. These, again, contribute to a better understanding of IAEA needs in specific areas and are beneficial in looking ahead for future research avenues to be eventually implemented in forthcoming years.

3.3 Collaboration with other Support Programmes

Given the organisation of the European Union and the existence of the ESARDA association – focusing on R&D for Safeguards, it is considered positive to disseminate JRC current R&D activities for the IAEA to other EU Member States with an active MSSP.

Ten EU Member States participate at IAEA's MSSP: Belgium, Czech Republic, Finland, France, Germany, Hungary, Netherlands, Spain, Sweden and the United Kingdom. These Member States are invited to participate at the EC-SP's Annual Review Meeting. In some cases, when discussing specific tasks or IAEA requests, it is beneficial to extend the discussion to other Support Programmes. This practice has been found useful both from the IAEA's perspective and from the participating MSSPs. Not only the discussions are richer, but also it is possible to better coordinate and focus on future efforts and initiatives.

Further to the above mentioned meetings, JRC researchers participate actively at ESARDA Working Groups. These working groups constitute a forum for technical discussions and contribute to a wide, scientific and technical knowledge base of Nuclear Safeguards. Participants to these working groups include ESARDA members and recognised observers. This is to say that both ENER and IAEA are represented in the working groups. As such, ESARDA working groups also contribute to the dissemination of the technical activities of many Support Programmes, including the EC-SP.

4. EC-SP Tasks

Since 1981, the EC-SP has been involved in as many as 117 tasks. Figure 1 shows the distribution of these tasks along the different Safeguards technical and application areas.

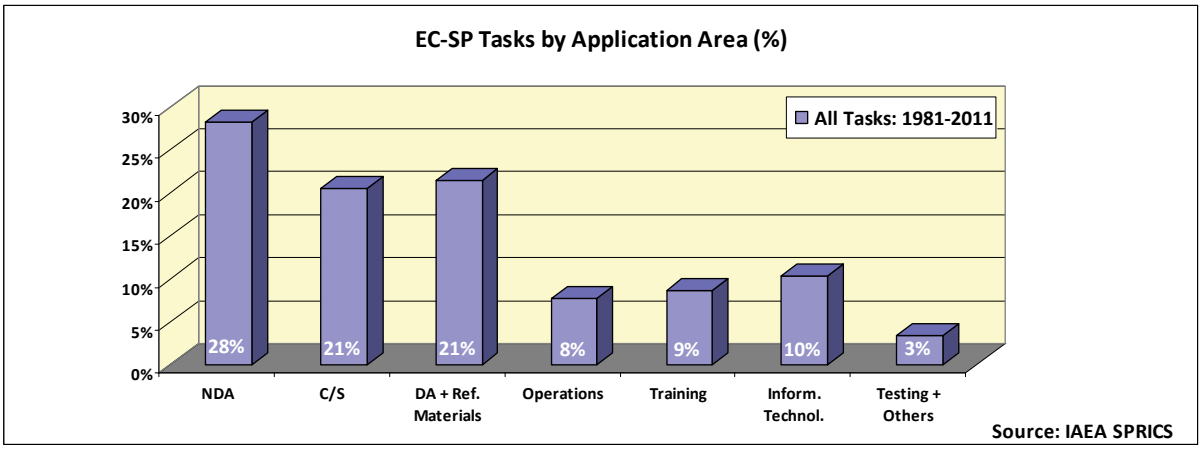

Figure 1: Distribution of EC-SP tasks along the different Safeguards technical and application areas for the period 1981-2011.

In Spring 2011, the situation of the European Commission's Support Programme is as follows:

NDA: Equipment, Modelling and Measurements	7
Sealing, Containment and Surveillance	8
Analytical and Reference Techniques	6
IAEA Operations (Ex. JNFL, JMOX Projects)	7
Information Technologies for Non-Proliferation	4
Training	7
Testing and Others	4
Total	43

Table 1: Distribution of EC-SP tasks in Spring 2011

Figure 2 shows the evolution of the EC-SP along the years in terms of the distribution of its tasks in terms of the different technical and application areas. The graph compares the distribution of all 74 closed tasks with the current 43 active ones. Figure 3 shows the number of active tasks since 1981.

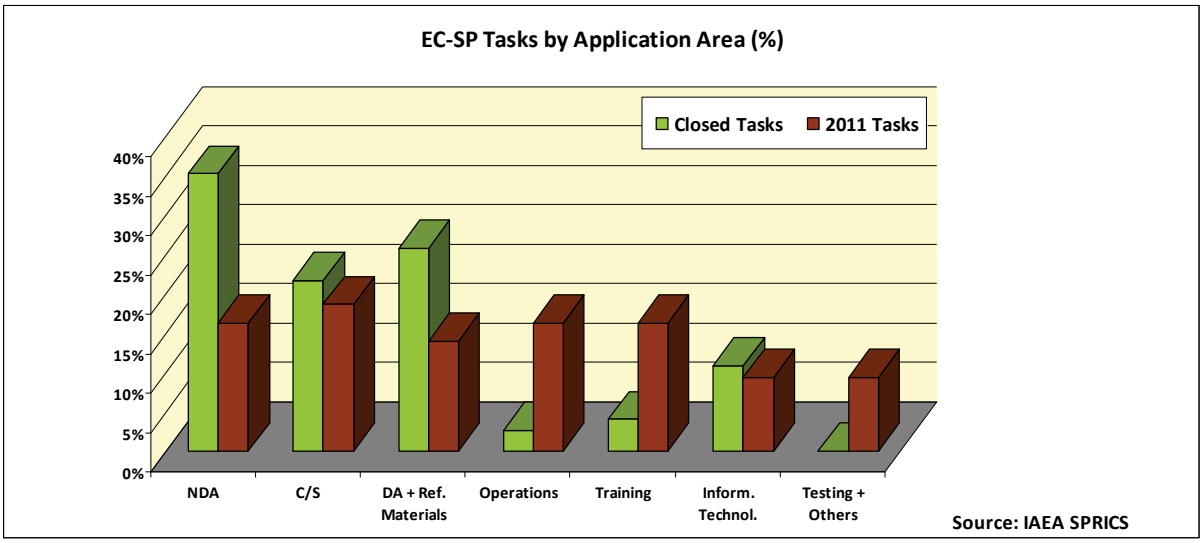

Figure 2: Distribution of EC-SP closed and active tasks in terms of the Safeguards technical and application areas

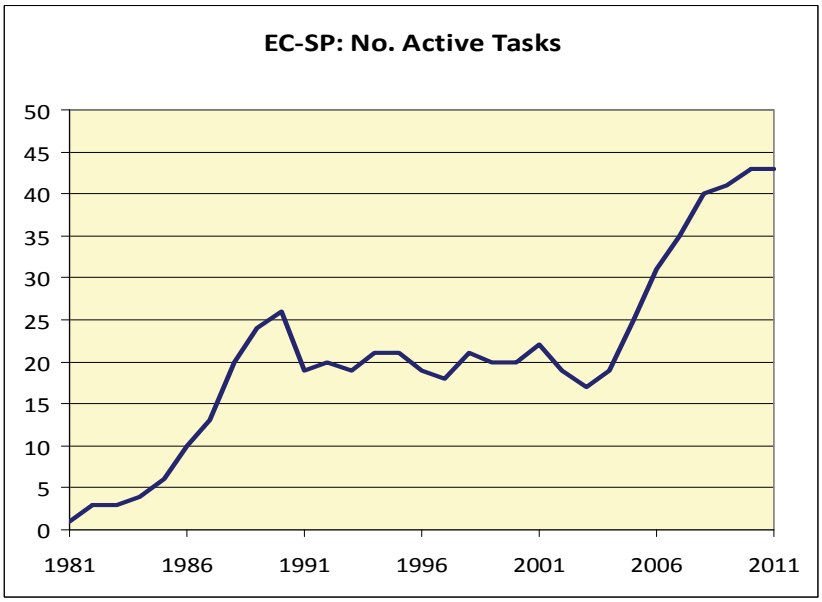

Figure 3: Number of EC-SP active tasks.

From Figure 2 the following is observed:

a) The relative weights of tasks associated to Containment and Surveillance (C/S) and Information Technologies are stable.

b) The relative weight of EC-SP tasks associated to traditional disciplines, such as NDA or DA, has decreased.

c) There is a substantial increase in tasks associated to IAEA operations and training.

d) EC-SP accepted IAEA requests in new activities (last column). Examples include: ASTOR Network of Experts for Safeguards in Geological Repositories, Novel Technologies and Safeguards by Design.

5. Recent Highlights of the EC-SP

This section lists a few recent EC-SP task highlights, illustrating how EC-SP developments can be close to inspectors' work and field measurements.

5.1 Reference Materials

The IAEA continues receiving reference materials and particles as requested. In particular, the use of the new uranium IRMM-3100a: 233U/235U/236U/238U=1/1/1/1 "quad" isotopic reference material was demonstrated [EC A 00318 – Special Reference and Source Materials for DA]. This new isotope reference material is ideal for verifying the inter-calibration of multi-detector systems in isotope mass spectrometry. As a result JRC-IRMM certified reference materials are now regularly applied in IAEA SGAS measurements strengthening the effectiveness and efficiency of IAEA Analytical Services.

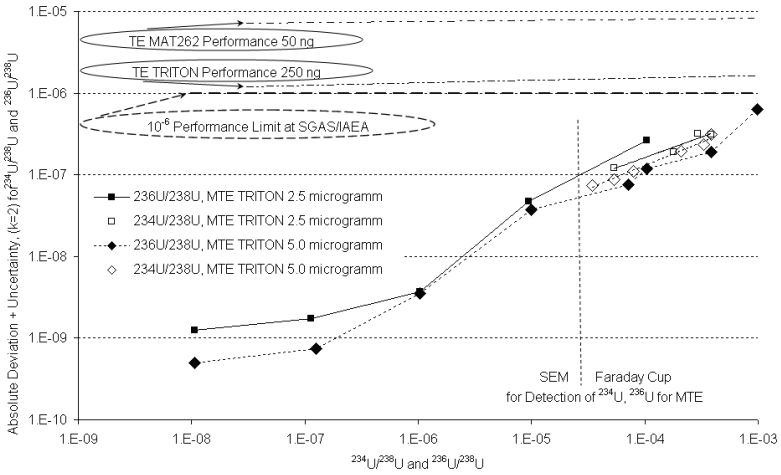

Figure 4: Performance of the Modified Total Evaporation method for minor uranium ratios n(^{234}U)/n(^{238}U) and n(^{236}U)/n(^{238}U)

Associated to the work in reference materials, advanced training techniques in Mass Spectroscopy were developed and experience transferred to the IAEA [EC B 01752 – Mass Spectrometry Training]. In particular, there were courses in thermal ionization mass spectrometry (TIMS). The capabilities and sample throughput for measurements of the minor uranium isotope ratios have been improved by implementing the so-called "modified total evaporation technique" at the IAEA Safeguards Analytical Services (SGAS-Seibersdorf). The performance of the MTE method for the minor uranium ratios n(^{234}U)/n(^{238}U) and n(^{236}U)/n(^{238}U) is seen in Figure 1. It is well below the performance limit for the entire range of ratios as required by the IAEA [1]. The excellent performance of this technique was demonstrated and it is now officially accepted for safeguards measurements at the IAEA SGAS. The MTE method was successfully integrated for routine use at IRMM, ITU, IAEA-SGAS and NBL and is applied for measurements of samples in safeguards and nuclear forensics.

In 2010 JRC-IRMM provided results to the IAEA on verification measurements of the recently domestic produced and certified JAEA LSD spikes and of batches of IAEA LSD spikes used for measurement of uranium and plutonium in fissile material control at the onsite laboratory in Rokkasho [EC A 1806 – Verification of mixed U-Pu Spikes] [2]. The reference materials used to accomplished this task were subject to a an inter-calibration campaign using state-of-the art measurement procedures carried out linking IRMM plutonium spike reference materials to underpin the confidence in the use of these IRMM reference materials for safeguards verification and environmental measurements. The compatibility of selected IRMM plutonium reference materials was demonstrated and the traceability of the certified values of the plutonium isotopic contents to the SI was confirmed. In addition this study was also linked to IRMM's successful participation in the external plutonium interlaboratory comparison programme EQRAIN-11, demonstrating IRMM's measurement capabilities for plutonium analysis [3].

5.2 Large Geometry Secondary Ion Mass Spectroscopy – LG-SIMS

The analysis of environmental particle samples is one of the means to detect the occurrence of undeclared activities dealing with enrichment and processing of nuclear materials. The techniques used today have proven to be effective for Safeguards measures and is a corner stone in the implementation of IAEA's additional protocol. For many years JRC-ITU has been involved with the development of novel analysis techniques aimed at the accurate identification of the constituents of fine particulate material. The ultimate goal is to perform accurate and precise measurements, determining the isotopic composition of the particles selected. This is of utmost relevance for safeguards as these particles are representative of the original material and their composition provides specific information about the source and, often, about the chemical/industrial processes used [4]. The results of this R&D effort are regularly communicated to the IAEA which, eventually, incorporates them as part of their standard analysis methodology and procedures.

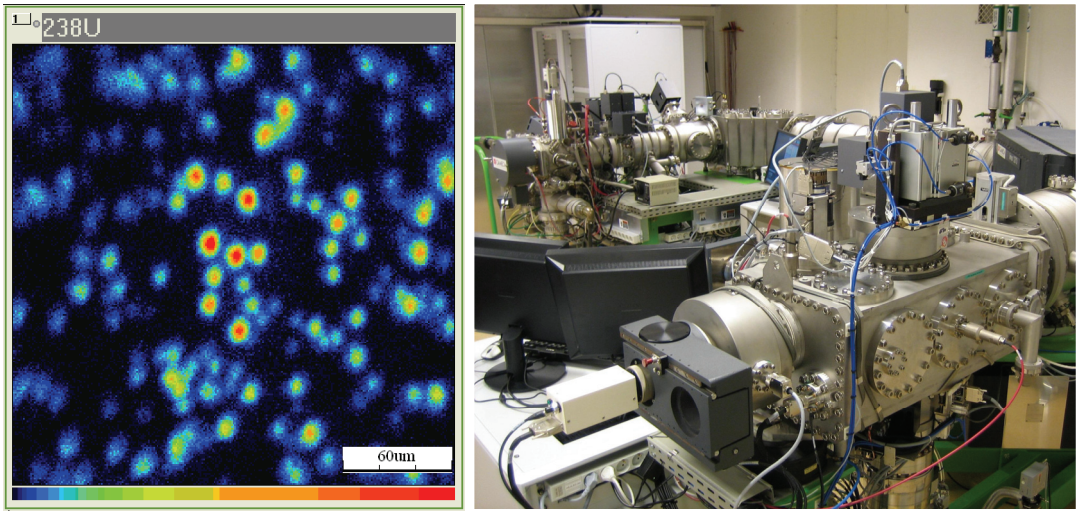

Figure 5: Example of JRC-ITU Automated Particle Measurement (APM) screening software that was recently developed by the company Cameca in cooperation with JRC-ITU, and a photo of an LG-SIMS from NORDSIM laboratory, Stockholm (equal to the one to be soon installed at JRC-ITU).

JRC-ITU is currently in the process of acquiring a Large Geometry Secondary Ion Mass Spectroscopy instrument – LG-SIMS. This instrument is the very same as the one that the IAEA will have in a near future as part of the new SAL laboratories. LG-SIMS improves the performance in uranium particle analysis, namely, high sensitivity at high mass resolution. Common molecular interferences are removed effortless, thus improving the minor isotope measurements. The results of uranium isotopic measurements are comparable to the best available TIMS measurements. The implementation of LG-SIMS will strengthen the Safeguards capabilities as it combines highest performance with a timeliness that does not exist today with the current use of small geometry SIMS and the fission track – TIMS method. This instrument also improves the detection capabilities of particles in a large matrix of other material.

5.3 COMPUCEA: Combined Procedure for Uranium Concentration and Enrichment Assay

COMPUCEA [Task EC A 01507] (Combined Procedure for Uranium Concentration and Enrichment Assay) is used for on-site analytical measurements in support of joint Euratom-IAEA inspections during physical inventory verification (PIV) campaigns in European Low-Enriched Uranium (LEU) fuel fabrication plants. The analytical technique involves the accurate determination of the uranium element content by energy-dispersive X-ray absorption edge spectrometry (L-edge densitometry) and of the 235U enrichment by gamma spectrometry with a LaBr3(Ce) detector. For evaluation of the LaBr3 spectra a modified version of the NaIGEM code is used, which has recently been adapted to handle the presence of reprocessed uranium.

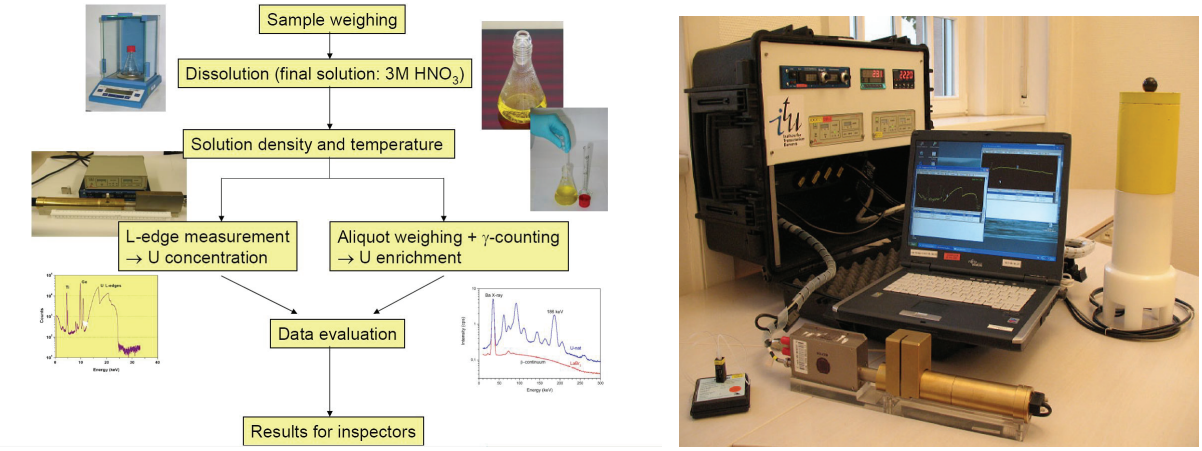

Figure 6: Procedure for the COMPUCEA technique and equipment

Following the successive and extensive evaluation of COMPUCEA's performance [5, 6], both in the laboratory and in field, the technique is now proposed to be used by the IAEA outside the European Union. First tests have already occurred and training actions are being prepared.

5.4 Ultrasonic Seals

JRC developed an ultrasonic seal [Task EC-E-01559] that is used in its different versions by EURATOM and IAEA Safeguards systems [7]. The internal structure of the ultrasonic seal comprises a unique non-reproducible identity and a frangible element (integrity) which breaks when an attempt is made to remove the seal from the sealed item. The reading device consists of a transducer which generates an ultrasonic signal and senses the reflected signal. The transducer rotates above the sealing bolt recording the ultrasonic echoes reflected over a complete revolution.

The core of the ultrasonic seal (photo to the very right, below) is a cylindrical assembly containing its unique identity and an integrity feature which breaks when opened. This assembly is radiation resistant and particularly reliable even under very harsh environmental conditions.

The identification feature is an assembly of several discs randomly stamped which are stacked in a random disposition and brazed together to form a univocal identity (second from left photo). Brazing paste is put in several parts of the stack in a quantity that will adequately braze the disks, but not fill all the holes. This is done by heating them up to 1000°C for several minutes in the furnace. As the diffusion of the brazing follows a random process, it is not possible to predict the identities that will be produced. The parts providing identity and integrity are then brazed together to form the core of the ultrasonic seals. This core is then welded into the top of the seal. The bodies of the seals are designed according to each application.

Figure 7: Core of ultrasonic seals.

Following the validation by an independent vulnerability assessment study, JRC ultrasonic seals are now classified as category A equipment and are used in nuclear installations in Romania, Canada and Pakistan.

5.5 3D Laser range Finder for Design Information Verification

Design Information Verification – DIV – is becoming increasingly important in International Safeguards as a way to verify that a plant set-up is consistent with the declared intent of its activities. Because of the complexity of nuclear installations, design verification can be extremely challenging and time consuming. Within the framework of Task EC-E-01425, JRC-IPSC developed a 3D laser based tool for Design Information Verification (DIV) purposes [8]. The DIV system [9] is divided in two main components: (i) a commercial off-the-shelf laser range scanner for data acquisition, i.e., capture the 3D coordinates, of a given environment with millimetre accuracy and (ii) a suite of JRC developed software applications needed to create an accurate 3D reference model, automatically analyse a verification model and detect changes, as well as manage all acquired and processed datasets, including secure storage and data authentication. This JRC developed system – 3DLR – is currently being used by the IAEA and ENER.

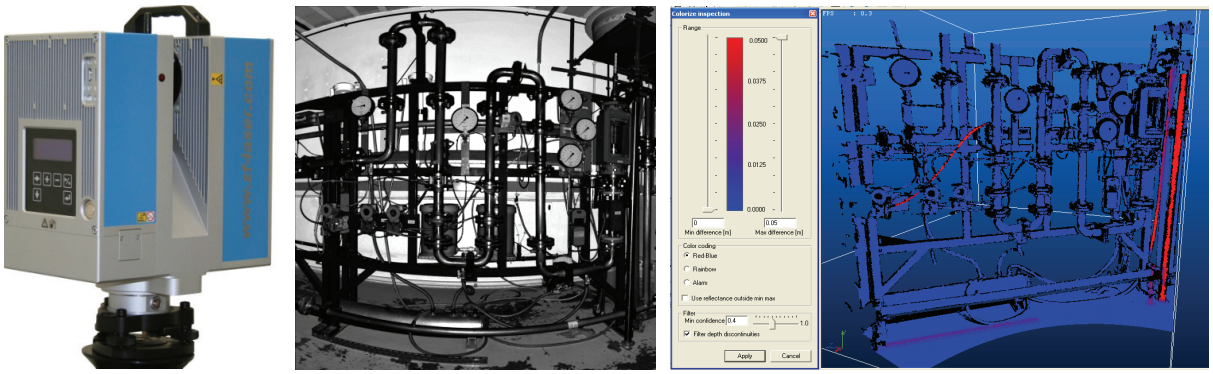

Figure 8: 3D Laser Range Finder used in the 3DLR, and examples of 3D capturing of a complex scene including automated 3D scene change detection – for verification purposes.

5.6 Development of an integrated approach to GCEP safeguards

The current IAEA strategy on safeguarding enrichment plants is still based on the results of the Hexapartite Working Group dating back to the early eighties. This working group tried to develop a system of mutually (IAEA and operators) acceptable assurances allowing the inspection of GCEP without disclosing sensitive technological information. Since then a lot of technological improvements have occurred, more countries had access to gas centrifuge technology and undeclared enrichment programmes have been discovered. All these factors call for an upgrade of the safeguards approach and the IAEA has started a process of modernisation of the inspection concept at GCEP's.

The current system relies on regular inspections for nuclear material accountancy based on NDA verifications (mostly on product cylinders) complemented by containment and surveillance measures. In addition LFUA (Limited Frequency Unannounced Access) to the cascade hall are allowed.

JRC is working in the frame of the EC-SP to develop an innovative integrated approach to GCEP safeguards that could improve effectiveness in the verifications of the kind of plants. The approach is based on three pillars:

- Continuous monitoring of load cells at feed/withdrawal stations, complemented with cascade modelling, aiming to a nearly real time accountancy (NRTA) of material in the plant
- Tracking/identification/authentication of cylinder flow in the plant
- Improved NDA techniques for verification of cylinders

The first goal is currently not fully addressed in an ongoing SP task, even though it is partly included in the proposal 10/TAU-005 "Evaluation of data collected from operator systems at enrichment plants" aiming to the analysis and evaluation of operator provided data at the GB-II plant. The general principle is to acquire continuously cylinder weight data from the load cells at the feed/product/tail

stations (mostly provided by operator equipment) and to analyse them in order to confirm the plant operation according to the expected behaviour and to exclude the presence of undeclared operations and/or the diversion of material. Since the monitoring is done only at the endpoints of the plant and no physical parameters are measured in the cascade hall, there is the need to develop theoretical models simulating the functionality and operation of the centrifuges and cascades in order to be able to analyse and correlate the signals measured at the entry and exit points and to conclude on the compliance of the operations with declarations.

The second part is done under task EC-E-1696 “L2IS: Laser Item Identification System” and aims to have a real-time tracking of flow of cylinders in the plant, complemented with identification and authentication features. The L2IS is capable of monitoring all transfers of UF6 cylinders between process areas by uniquely identifying each cylinder through exploring the unique 3D microstructure of each cylinder’s surface. It has been demonstrated that every cylinder has a unique ‘fingerprint’ which remains stable even under extreme environmental conditions. The L2IS system is composed of a portable unit, operated in attended mode, and a fixed installed unit, operated without inspector presence. An inspector, using the portable unit, acquires the fingerprints of a given set of feed cylinders intended to be used over the forthcoming months. The fixed system monitors the flow of previously identified cylinders in a transfer corridor. This system is coupled with standard video surveillance that can remotely transmit state of health information to IAEA Headquarters. The video surveillance can be interfaced with electronic seals applied to the cylinders to record and display seals data (e.g. status, time/date of application). The integration of data from the L2IS with data from weighing and NDA stations is foreseen to monitor and verify all transfers. This will provide a high deterrence of diversion or substitution, and an increased probability of detection thereof. After one year of field testing, successful results of the L2IS have been reported [10].

Figure 9: L2IS Portable and Fixed reading Stations

Finally the improved NDA on cylinders is executed under task EC-A-1687 “State of the Art of NDA Techniques Applicable to UF6 Cylinders”. The current verification system on cylinders relies on accurate weight measurements at the accountancy scales and on gamma spectrometry for enrichment measurements. Current technology on gamma spectrometry does not allow to reach the wished accuracy in the cylinders used at GCEP plants: the large cylinder wall attenuates too much the soft X-rays needed to perform spectral analysis with intrinsic calibration methods and the measurements done based on the enrichment-meter principle require an accurate knowledge of the wall thickness in order to correct accurately for the attenuation [11]. JRC has analysed the potentiality to improve the measurements of cylinders using passive neutron measurements. This alternative technique is based on the measurement in a well controlled geometrical configuration of the total neutron source generated by (α,n) reactions within UF6. This is not a direct indicator of enrichment since the main contribution to the neutron source comes from 234U. Nevertheless 234U/235U ratio is constant within a plant and known when the enriched UF6 is directly produced from natural feed, which is the most common operational case. The application of the technique could be problematic to cases such as blending of products, reprocessed uranium, re-enrichment of tails, products from enriched feed.

6. Discussion and Conclusions

JRC's experience in operating the European Commission Support Programme, in line with the collaboration with ENER, has been very positive. The franc dialogue with the IAEA led to a programme of applied research targeted to Nuclear Safeguards applications. This programme has produced several pieces of work with relevance to International Safeguards stakeholders.

In recent years, it is possible to say that EC-SP contributions, while maintaining activities in research and development in Nuclear Safeguards basic disciplines – C/S, DA and NDA, have increased in the areas of operations and training. This is the natural evolution of product development, i.e., passing from laboratory prototypes to dedicated field instruments and measurement systems.

The EC-SP has kept in line with the new orientation of the IAEA in having "Safeguards which is fully Information Driven". Indeed, in the last six years there have a been a few tasks paving the way and exploring new ways to acquire, process, analyse and integrate multi-lingual, multi-source, multi-timeframe information, including trade data.

In a domain as technical as Nuclear Safeguards, with a constant evolution of equipment and methods, training plays an important role to keep IAEA staff abreast of the new developments. Within the framework of the EC-SP, and for the last 30 years, JRC has made available its installations, laboratories, materials, expertise and know-how to the IAEA. There are tasks associated to long-standing training needs. Besides those tasks, other tasks often include a dedicated component of training, associated to the specific topic of the task.

The existence of a Support Programme creates, somehow, a sense of partial ownership in what concerns the implementation of International Safeguards. This makes politicians and decision makers more informed about IAEA Safeguards, its rules, modes of operation and technical requirements. This is specifically true for all the scientific and technical staff working in JRC laboratories who feel most gratified when they know that their work has successfully contributed to the continuous challenge in "raising the bar".

Thirty years is a long period. The European Commission Cooperative Support Programme feels proud for all its past activities and achievements. The EC-SP wishes that the next thirty years are as successful and looks forward to increasing cooperation with the IAEA and its Member States Support Programmes.

7. Acknowledgments

The author expresses his gratitude to all colleagues in ENER and JRC for their collaboration in the smooth implementation of the European Commission Cooperative Support Programme. In particular, a special word for all JRC task officers and other scientific and technical staff who play an important role in the execution of the different tasks as well as for having provided material for this paper.

A special acknowledgement is due to the previous EC-SP Coordinators Messrs. Marc Cuypers and Sergio Guardini who helped tracing back part of the history of the EC-SP.

The author also acknowledges IAEA's Support Programme Coordination team for the excellent relations and good discussions aimed at the continuous improvement and efficiency of the EC-SP.

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Transparency and other State-Specific Factors: Exploration of Ideas for Evolving the IAEA's System of State-Evaluations and Safeguards Implementation

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Abstract:

In November 2010 the IAEA Department of Safeguards launched its Long Term Strategic Plan at the IAEA Symposium on International Safeguards: 'Preparing for Future Verification Challenges'. A key element of the Long Term Strategic Plan is the further evolution of the State-level approach for safeguards implementation away from criteria-driven safeguards approaches focussed at the facility level, to a safeguards system that is objectives-based and fully information-driven. The State-level approach is a holistic approach to safeguards implementation, applicable to all States, incorporating comprehensive State evaluations and safeguards implementation approaches that make use of all information available to the IAEA.

In further evolving the State-level concept State-specific factors and acquisition path analysis will become increasingly important in State evaluations and in the determination of safeguards approaches for each State. It will be important to determine objective modalities for incorporating these factors. Consideration of State-specific factors in determining safeguards approaches is not new – in fact, paragraph 81 of INFCIRC/153 (concluded June 1972) enumerates several such factors that can be considered. This paper will explore some ideas for State-specific factors that could be used in State-evaluations, and how these factors could be used for determining State-by-State safeguards approaches. Ideas for State-specific factors will include effectiveness of State Systems of Accountancy and Control (SSAC), transparency of States in their dealings with the IAEA, and characteristics of a State's nuclear fuel cycle.

Keywords: State-level concept, safeguards implementation, State Systems of Accountancy and Control

1. Introduction

In November 2010 at the International Atomic Energy Agency (IAEA) Safeguards Symposium, the Deputy Director-General for Safeguards, Mr Herman Nackaerts, launched the Department of Safeguards Long Term Strategic Plan (LTSP): 'Preparing for Future Verification Challenges'. A major focus of the LTSP is the further evolution of the IAEA's safeguards system away from criteria-driven safeguards approaches[i] focussed at the facility level, to a safeguards system that is **fully information-driven** – in other words, making greater use of **State-level approaches** that utilise all information available to the IAEA about the State, both facility-specific factors and State-specific factors. The characterisation "further evolution" is important here, as the State-level approach to safeguards implementation is not new, rather it has been used to varying degrees for several years, and its pedigree derives from several provisions in safeguards agreements and guidelines going back several decades.

Examples of types of information used under a State-level approach include, inter alia: level of cooperation by the State; effectiveness of the SSAC (State System of Accounting for and Controlling nuclear materials); characteristics of the State's nuclear fuel cycle, etc. Some established de jure and de facto bases for a State-level approach are listed below:

INFCIRC/153, para 3 (1972): ...the Agency and the State shall co-operate to facilitate the implementation of the safeguards provided therein

INFCIRC/153, para 81 (1972): ... criteria to be used for determining the actual number, intensity, duration, timing and mode of routine inspections of any facility shall include:

- the effectiveness of the State's accounting and control system ...
- the extent to which the operators of facilities are functionally independent of the State's accounting and control system
- the extent to which an appropriate accountancy and control system is in place [following measures specified in INFCIRC/153, para 32] for each material balance area is implemented by the State
- the promptness of reports submitted to the Agency
- the characteristics of the State's fuel cycle
- international interdependence of the State's nuclear fuel cycle

IAEA/SG/INF/2, Forward (1980): The following factors are considered of primary importance with respect to the effectiveness and credibility of IAEA safeguards:

- cooperation between the Agency, the State and the facility operator in implementing the safeguards
- the adequacy of the SSAC in relation to IAEA requirements for accounting for and control of nuclear material

GOV/2002/8: The Conceptual Framework for Integrated Safeguards (GOV/2002/8) described the importance of information review and evaluation as a fundamental element of strengthened and integrated safeguards. The implementation of integrated safeguards by the Agency, following the conceptual framework, was endorsed by the Board of Governors in early 2002.

Paragraph 3 of INFCIRC/153 is worthy of greater exploration with respect to one of the key State-specific factors, transparency – an important aspect of cooperation. The fact that INFCIRC/153 states the importance of cooperation so early in the document demonstrates it is an important foundational principle to safeguards implementation. The negotiating records of INFCIRC/153 reinforce this, noting that the fact that there is an early paragraph in INFCIRC/153 on the importance of cooperation, and that this come before the paragraphs that elaborate restraints on the IAEA in implementing safeguards (paragraphs 4-6), demonstrates the priority of cooperation in the eyes of the drafters[ii].

There are also some very practical reasons for the IAEA to make use of a State-level approach. The IAEA has an obligation to provide credible assurances to the international community that states' nuclear activities are for peaceful purposes[iii]. It must do this within its allocated budget and resources which are limited and are likely to remain so as nuclear activities expand around the world in the coming years. This is elaborated in paragraph 6 of INFCIRC/153 which states that the IAEA must "make every effort to ensure optimum cost-effectiveness ...". In a world where the quantity of safeguarded nuclear material is increasing as the nuclear industry expands, if the IAEA were to simply follow mechanistic approaches based on the quantities and types of nuclear material and facilities in States then it simply could not do so within a constrained budget without reducing the confidence of its verification conclusions. This would clearly not be an acceptable state of affairs. However, using a holistic approach to safeguards evaluations that makes greater use of <u>all</u> information of safeguards significance the IAEA holds on a State, and uses the evaluation of that information to determine appropriate safeguards implementation measures, has the benefit of potentially significant efficiency gains, without affecting safeguards effectiveness.

2. State Evaluation Processes

The key elements to the IAEA's State evaluation processes are the: State Evaluation Report (SER); State-specific Implementation Plan; and the State-Level Approach (SLA). Collectively these can be referred to as the State Evaluation process and, implemented together iteratively, provide the foundation of the safeguards system that is fully information-driven.

A State Evaluation is a comprehensive analysis of all the information available to the IAEA (from all sources) on a State's nuclear program, and is designed to provide the IAEA with a thorough understanding of a State's nuclear and nuclear-related activities. It is the State Evaluation process that provides the context against which safeguards resource allocation decisions are taken. The search for indications of proliferation-related activities is a key element of the State Evaluation process. One of the most important means of detecting signs of covert nuclear activities is the identification of inconsistencies in declared and other relevant data. The identification of inconsistencies in the data requires careful analysis and matching of data from a variety of sources. In this regard, it is important that the information-driven safeguards net be cast widely.

Arising from the SER process the IAEA produces a State Level Approach to safeguards that applies the IAEA's understanding of the State's nuclear fuel cycle to the safeguardable activities in the State, to produce an approach that ensures that appropriate safeguards measures are in place to address possible diversion scenarios and to enable the IAEA to draw credible conclusions that there is no undeclared materials or activities of safeguards significance taking place within the State.

The State-Specific Implementation Plan is the practical expression of the State Level Approach. It includes when, where and how safeguards resources are to be allocated. On the basis of the Implementation Plan, safeguards inspections, complementary accesses, design information verification visits and headquarters activities are scheduled and assigned.

The output of the activities conducted under the Implementation Plan is the reports produced by the inspectors which feed into the safeguards conclusions for the State. These safeguards conclusions feed into the SER, which can result in modifications to the SLA which can in turn lead to changes in the Implementation Plan. This process of review, refinement, and change leads to a form of safeguards that is adaptive to change, responsive to facts on the ground and, in time, fully information driven.

It is important to note that State-specific factors have been taken into account for some time by the IAEA in its State Evaluation processes. However, the important point with regards to evolving the IAEA's safeguards system is that consideration of State-specific factors has not generally led to changes in safeguards Implementation Plans for each State. In other words, consideration of State-specific factors has influenced the IAEA's safeguards conclusions on States, but generally has not influenced the frequency, intensity and scope of in-field inspection activities. It is the move to combining State Evaluation processes and in-field inspection-related activities that is key to the evolution of the Safeguards System.

3. What should drive the State Evaluation process?

The following provides a snap shot of some State-specific factors that can be taken into account when performing a State Evaluation[iv]. This is not an exhaustive list, and much, if not all of this is already taken into account by the IAEA to some degree, but factors such as those listed here will take a greater importance in State Evaluations as the IAEA evolves its safeguards system to make fuller use of State-level information. In the list below, where a State-specific factor is one that is included in paragraph 81 of INFCIRC/153 as a "criteria to be used for determining the actual number, intensity, duration, timing and mode of routine inspections", it is referenced as such.

Making use of State-specific factors is not a case of discriminating between States with equivalent safeguards obligations, rather it uses objective technical criteria to differentiate between states in the safeguards measures applied. As such, it is important that there is consistency between one State and another in the State Evaluation process and modalities used, with the differentiation arising from

how each State measures up against each State-specific factor. Some of the factors listed below are amenable to quantitative differentiation between States, whilst others are more judgement evaluations. The challenge is putting together a range of information about a State, some quantitative and some qualitative, to make a judgement on differentiating between one State and another with how safeguards is implemented.

3.1. History of acceptance of non-proliferation norms

A State's history of accepting non-proliferation norms can include: adherence to the NPT, IAEA Safeguards Agreements, and the Additional Protocol; established national policies in support of non-proliferation, backed up by robust legislation, etc. If these factors are complemented by a history of the IAEA drawing positive safeguards conclusions on the State then this could form a strong State-specific factor. This has the additional advantage of being semi-quantifiable, hence amenable to quantitative differentiation between States.

3.2. SSAC Effectiveness

The effectiveness of a State System of Accountancy and Control (SSAC) is clearly an important factor (and is listed as such in paragraph 81(a) of INFCIRC/153) but evaluating the effectiveness is not as readily amenable to quantitative differentiation between States. There is a reasonable level of judgement in this factor, but experienced safeguards inspectors that have dealt with many different SSACs, can differentiate between a poor performing and high-performing SSAC – i.e. SSACs with which the IAEA has confidence have the requisite regulatory authority and culture with which to ensure safeguards compliance in their country. There are some measurable structural elements in the suite of information that contributes to an SSAC's effectiveness, such as: regulatory independence from facilities [153/81(b)]; enforcement powers; accountability to national parliaments. There are also measurable elements in relation to the SSAC's record – e.g. correctness and timeliness of reports.

3.3. SSAC Cooperation and Transparency

This is likewise difficult to evaluate in a way that enables quantitative differentiation between States. Cooperation and transparency can manifest themselves in both a macro and micro way. Important macro factors could be, for example, the extent to which a State's nuclear fuel cycle is interdependent with other States. Paragraph 81(d) of INFCIRC/153 lists this in the context of an interdependence of States for receiving and sending nuclear material, but this can be extended to interdependence of fuel cycle processes. For example if a State has a nuclear fuel cycle plant that is run as a multi-lateral consortium of countries that are all of good non-proliferation standing. Some examples of micro-factors might include the day-to-day responsiveness of the SSAC to IAEA questions and enquiries, the flexibility the SSAC applies in giving the IAEA access to sites and information, etc.

3.4. Fuel Cycle Rationale

Fuel cycle activities in the state should have a clear rationale related to the commercial, energy or scientific needs of the state and its known trading partners. This is especially the case for proliferation-sensitive developments, i.e., activities related to isotopic separation or plutonium extraction. Given the direct relevance of these technologies to proliferation and the availability worldwide of enrichment and reprocessing services from commercial providers, the rationale for a State to be developing such capabilities would warrant very close scrutiny in the context of the IAEA's State Evaluation process.

3.5. Coherency

An underlying principle in a holistic assessment of a State's nuclear program is that the program should fit together to form a coherent whole where each activity can be placed in context with clear relationships to other parts of the program. This includes:

- checking that declared nuclear activities fit together within the State's civil program as a whole, or match an established or prospective pattern of trade;
- identifying questions and inconsistencies that require further investigation; and
- the identification of possible indicators of undeclared nuclear activities.

Any activities which do not fit within this coherent pattern may indicate the possibility of undeclared activities, and as such warrant closer attention in the IAEA's State Evaluation process.

3.6. Consistency

For States with an Additional Protocol in force the IAEA has the benefit of access to a range of nuclear-fuel-cycle-related information that it does not receive from States with only a comprehensive safeguards agreement. As such, for these States, the IAEA's State Evaluation process can include the following consistency checks in relation to nuclear-fuel-cycle-related activities:

- internal consistency of the Additional Protocol Declaration;
- consistency of Additional Protocol Declaration with information obtained via environmental sampling, open source information, trade information, etc;
- consistency of the State's declaration of exports and imports of nuclear, nuclear-related material and equipment, etc., with other states' declarations.

An inconsistency should not automatically be given prominence in the State Evaluation process; rather the IAEA needs to evaluate the significance of any inconsistency in deciding what follow-up actions are warranted. Inconsistencies may be an indicator of undeclared activity, or an innocent mistake based on erroneous declarations, or a misunderstanding on the part of the IAEA.

3.7. Nuclear Material Flows

Paragraph 29 of INFCIRC/153 states that nuclear material accountancy is a "safeguards measure of fundamental importance". This is an important point, as an evolution to a safeguards system that makes greater use of State-specific factors does not mean the IAEA would stop using nuclear material accountancy as a part of its verification toolkit. Data on nuclear material flows within an MBA (material balance area) is a facility-specific factor, not a State-specific factor, but the interpretation of this data in the context of the State as a whole can be considered a State-specific factor. A careful analysis of nuclear material flows through the State as a whole needs to be an essential component of State Evaluations. Where there are apparent anomalies in declared flows, or facilities exist with greater capacity than the declared throughput, this could be something that the IAEA needs to investigate further, however it should be done in the context of the State-level approach.

4. Putting it all together

Criteria-based safeguards approaches that mechanistically set the frequency and scope of in-field safeguards verification activities based on set detection probabilities, quantity and timeliness goals, are by their very nature amenable to quantitative analysis and comparison between States, so can be appealing from an analytical-perspective. The rigid application of these approaches however do not readily accommodate consideration of all information on a State in setting in-field activities, and in practice can lead to the use of inefficient in-field safeguards verification activities beyond what is necessary to achieve the required safeguards effectiveness. Conversely, drawing more broadly on both nuclear material accountancy and facility information and State-specific factors allows the IAEA to objectively and flexibly consider the full suite of information available on a State in setting in-field safeguards verification activities. This approach however does have the complexity of having to deal with a mix of qualitative and quantitative data that is less amenable to quantitative analysis and comparison between States.

Following a State-level approach will lead to variations from one State to another in the intensity, frequency and scope of in-field verification activities. As such, the IAEA will need to be able to justify and explain those actions as being a differentiation of approaches, not discrimination. To do so, the IAEA's State-by-State decisions on setting safeguards verification activities will need to be based on analytical arguments that are transparent to Member States. Accordingly, it will be important for the IAEA to be able demonstrate that it employs a consistent, objective methodology for all States.

So, what consistent, objective methodologies could the IAEA use for evaluating States and setting inspection-related activities? There are for example various discriminant analysis tools used in other sectors for evaluating multivariate quantitative variables and mixtures of qualitative and quantitative variables; such as in finance, shape recognition, and signal detection in noisy datasets used in experimental nuclear and particle physics. While many of the State-specific factors listed above are qualitative in nature, many comprise quantifiable elements that could, in principle, be combined into an overall numeric figure of merit.

Determining which analytical tools might be fit for IAEA safeguards evaluation purposes would be a substantial project, and is beyond the scope of this paper. One potential risk however is that using analytical statistical tools could simply continue the use of inflexible mechanistic approaches – albeit different ones – which is exactly what the IAEA is trying to avoid by moving from the criteria-based approach. It will be important in some way to maintain the IAEA's flexibility to apply professional judgement in assessing the relevance of the suite of State-level factors. Accordingly, if used, analytical statistical tools should not just be developed in the abstract, rather it would be important for the tools to be "road tested" to ensure they lead to reasonable conclusions and do not subjugate the role of professional judgement.

As an alternative to using complex analytical techniques to analyse mixtures of qualitative and quantitative data, relying on professional judgement could instead be a useful tool in the hands of experienced safeguards inspectors and analysts; it is just a question of ensuring that the use of professional judgement follows a consistent and objective process for all States. To use the professional judgement of only one person would run the risk of bias and inconsistencies between evaluations, and would probably not meet the test of demonstrating a consistent and objective approach for safeguards evaluations of all States. A more consistent way would be to aggregate professional judgement across several professional assessors by using a team of safeguards inspectors and analysts to conduct State Evaluations and to determine State-level approaches to safeguards implementation. The IAEA has implemented just such a process through the use of State Evaluation teams comprising up to about five people for each State that will meet throughout the year to evaluate States and to use these evaluations to determine appropriate safeguards implementation approaches.

5. Conclusion

The concept of the IAEA using State-level factors in safeguards evaluations is not new. Various State-level factors are enumerated in paragraph 81 of INFCIRC/153 and State-level factors have been used by the IAEA for around ten years in its implementation of Integrated Safeguards. But while State-level factors have been used in State evaluations, where they have had limited use is in determining the frequency, intensity and scope of in-field inspection activities. Making greater use of State-level factors will lead to changes in the frequency, intensity and scope of in-field inspection activities, even between States that have similar fuel cycles, but this will be a matter of differentiation on the basis of objective evaluations of States as a whole, not discrimination. In communicating the safeguards approaches to Member States it will be important for the IAEA to be able to demonstrate it is using consistent and objective approaches for all States.

This paper provided a representative mix of some State-level factors that could be used by the IAEA in applying the State-level approach. Some of these factors were quantitative in nature while others were more a matter of professional judgement (qualitative). A difficult challenge will be determining how best to combine mixtures of quantitative and qualitative factors in such a way as to make objective judgements on appropriate safeguards approaches to apply on a State-by-State basis. There are analytical statistical tools used in other sectors that could perhaps be applied to this safeguards problem, that have been discussed in this paper. Alternatively, the IAEA could make use of the aggregated professional judgement of a group of inspectors and analysts, along the lines of what the IAEA has begun to use with the establishment of State evaluation teams. Whichever approach is used, or a combination of the two, it will be important for it to be demonstrably consistent for all States, and to maintain an important role for professional judgement.

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iii. This obligation is outlined in paragraph 2 of INFCIRC/153. A typical expression of paragraph 2 in a comprehensive safeguards agreement is: “The Agency shall have the right and the obligation to ensure that safeguards will be applied ... on all source or special fissionable material in all peaceful nuclear activities within the territory of ..., under its jurisdiction or carried out under its control anywhere, for the exclusive purpose of verifying that such material is not diverted to nuclear weapons or other nuclear explosive devices.”

iv. See also: Frazar S. and Mladineo S. *Safeguards Culture: Lessons Learned*, ESARDA BULLETIN, No. 44, June 2010

Technical Implementation of Nuclear Nonproliferation Cooperation to Complement IAEA Safeguards

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Abstract:

Compliance with the terms of a state's international safeguards agreement with the International Atomic Energy Agency (IAEA) has long been a key underpinning of the nuclear non-proliferation regime. The framework for international safeguards relies on an independent organization, the IAEA, acting under bilateral agreements with states to verify declarations of nuclear materials and activities, and reporting its conclusions annually to the world at large. The system has worked well for decades; nevertheless, there has been and continues to be interest in supplementing the compliance-based system with voluntary measures by a state to demonstrate commitment to nuclear non-proliferation goals. Direct state-to-state exchange of non-proliferation-relevant information is one such approach. For example, the cooperation of Brazil and Argentina was an enabling factor in the accession of those two countries to the Treaty on the Non-proliferation of Nuclear Weapons (NPT) and acceptance of IAEA Safeguards. In some cases, a regional sharing of non-proliferation relevant information may be appropriate. Such cooperation has been of interest to various states in the Asia-Pacific in particular. The step from a bilateral exchange to multilateral cooperation presents a major technical challenge, however. To begin, interested parties to regional voluntary cooperation or "transparency" must clearly define the goals and expectations of the cooperation. There must be perceived benefit to such cooperation to justify the effort and costs, but at a minimum, the sharing must cause no harm. It is critical to establish the associated requirements for information sharing, which necessarily must address security concerns. Fundamentally, each party to the cooperation must be able to trust the information obtained from the regional network, and similarly, be confident in the security of information it discloses to the network. If implemented properly, a regional system can withstand the ups and downs in political relations between states. Measures for authentication and encryption of information are only part of any technical framework, which must be augmented with appropriate procedural measures as part of a system solution. Sandia National Laboratories has been a partner with Japan, Korea, the European Commission, and other states under bilateral agreements with the U.S. Department of Energy to develop sound technical approaches that facilitate regional non-proliferation cooperation.

Keywords: safeguards, regional cooperation, transparency, information security

1. Introduction

1.1. Safeguards by an independent, trusted third party: IAEA

Even before the appearance of the Treaty on the Nonproliferation of Nuclear Weapons (NPT), nuclear supplier states recognized the value of an independent third party to verify peaceful use of technology and materials transferred to recipient states. Accordingly, the International Atomic Energy Agency (IAEA) quickly assumed such responsibilities, which were codified under "project agreements" that implemented safeguards as described in IAEA Information Circular (InfCirc) 66. This third-party role of the IAEA in providing safeguards was instrumental in relieving states of what otherwise would have required an enduring bilateral relationship between supplier and recipient states. The ensuing duplication of effort, lack of uniform approach, lack of integration, overlapping jurisdictions, etc. would have led quickly to a costly, unmanageable, and unsustainable mess. With the advent of InfCirc 153 comprehensive safeguards agreements following the NPT, the IAEA safeguards system gained even better efficiencies through consolidation. In sum, the creation of this centralized framework for international nuclear safeguards was indeed instrumental to the success of the

nonproliferation regime. Much of the evolution of that regime has focused on what tools are necessary for the IAEA to implement safeguards effectively.

1.2. IAEA releases a safeguards conclusion, but not the data

While the creation of the IAEA safeguards system relieved states of direct responsibility for verifying the peaceful use of nuclear materials and technologies that had been shared with others, it did not dismiss their interest in the "answers." The IAEA announces a Safeguards Implementation Report (SIR) annually, which is limited to the conclusion drawn by the IAEA in respect of the implementation of safeguards in a particular state having a safeguards agreement with the IAEA. Information supporting the IAEA conclusion is not released, but instead is protected as "Safeguards Confidential." The confidentiality of safeguards information, an IAEA pledge to an inspected state, is argued as being necessary to ensure the full cooperation of the state with the IAEA. An inspected state could otherwise be reluctant to be completely forthcoming with the IAEA. Compliance information could entail proprietary secrets, reveal physical protection measures, or otherwise put at risk justifiable activities or assets if disclosed indiscriminately. Thus "safeguards confidential" is standard practice; it is a critical enabling factor for IAEA safeguards.

1.3. State transparency can complement Safeguards

For various reasons, states may at times desire more than the safeguards assurance provided by the IAEA. The global system is not meant to address all of the particular questions that might arise in limited geopolitical situations. The cooperation of Brazil and Argentina, which led to the formation of the Brazilian-Argentine Agency for Accounting and Control of Nuclear Materials, ABACC, is a good historical example. Direct cooperation was the enabling factor for both countries to accede to the Treaty on the Non-proliferation of Nuclear Weapons (NPT) and only then accept IAEA Safeguards. Twenty years later, ABACC still receives enduring support from the two states.

More recently, both Japan and the Republic of Korea have been exploring cooperation on the voluntary exchange of information related to their nuclear activities. In a workshop on regional transparency in Tokyo in February 2008, Wan Ki Yoon (Korea Institute of Nonproliferation and Control) described how the direct cooperation of states strengthened the nonproliferation regime.[1] He used the metaphor of a cone: The safeguards agreements of the IAEA, at the apex, comprised the sides of the cone, with individual sovereign states arrayed in a circular ring at its base. "Transparency" appears in the base of the cone, as an overlapping network of connecting lines between individual states. The complementary nature of compliance with IAEA safeguards and voluntary state transparency contributes to a more robust nonproliferation assurance.

Safeguards confidentiality is an obligation of the IAEA, not an obligation of the providing state. If a state should decide that sharing information with other recipients—even safeguards information—is in its national interest, there is nothing to prevent it from doing so. Such transparency carries both benefits and risks. Precisely *how* information might be shared for regional transparency applications can affect the balance of risks and benefits significantly. The purpose of this paper is to frame the problem for information sharing from a technical perspective.

2. Context for Nuclear Nonproliferation Cooperation

Unlike compliance with IAEA safeguards, voluntary information sharing between states is not governed by clear cut guidance on *what* information should be involved, nor *how* it should be shared. Many factors must be considered: the goals in sharing information, the types of information involved, what sort of reciprocity may be expected, associated threats and risks, the need to be able to trust the information, a scalable technical architecture for sharing, metrics to evaluate the viability and utility of a technical sharing solution, procedural resolution of anomalies, and a host of additional factors.

2.1. Different approaches to transparency

Before considering any technical implementation, it is important to consider the various ways a state might choose to provide nuclear nonproliferation information voluntarily. The following description is meant only to illustrate the differing contexts for consequent

technical solutions, rather than to discuss the relative merits of transparency approaches.

An option that is always open is unilateral information sharing. This could be completely open release, such as posting on a universally-accessible web site. On the other hand, unilateral sharing could instead be confined to some limited audience. Ideally, a "limited audience" should mean a single recipient, so that if the information were to appear elsewhere, it's clear who forwarded it. If more than a single recipient, there is little practical difference from open release without some effective mechanism that constrains further information sharing and dissemination.

Instead of using a unilateral approach, a state wishing to improve transparency could do so by working out an agreement with a recipient party. Although there is no reason why such an agreement couldn't involve one-way information sharing, it more likely involves reciprocal, two-way information sharing. The simplest situation is under a bilateral arrangement, with a single, consolidated channel for the associated information exchange.

Extending beyond a bilateral agreement, information sharing can be enlarged to multilateral involvement. One possibility might include a third party as an intermediary to facilitate an otherwise more difficult bilateral relationship. Or it could involve additional states all as relatively equal participants to a common arrangement. As soon as the context moves from bilateral to multilateral—the addition of just a third party—the implications for the technical dimension of information sharing become fundamentally more complicated. Indeed, technical solutions that sufficed for bilateral information sharing may no longer be acceptable, unless the original bilateral technical approach anticipated the scaling to multilateral from the beginning.

2.2. Considerations in "supplying" transparency

Here we assume that providing information is something that a state *wants* to do. It is not something that is required, or that has predefined requirements: it is entirely up to the state to decide how it would be done.

Most important, there must be a need that answers the question "why" it should be transparent. The need should be defined in clearly stated goals and objectives. It might be to address an explicit external request, respond to a real or perceived need, or just to provide assurance. In the nuclear nonproliferation realm, it is typically to demonstrate responsible stewardship of nuclear materials and technology: peaceful use, peaceful intent, competence of staff, etc. Lines can blur, particularly with transparency efforts related to operational safety, physical security, and related concerns.

A second critical consideration is to define precisely the audience: *Who* is the intended recipient of provided information? Presumably the state has identified a "need to know." At this stage, the state also may wish to consider who does not need to know, and why not.

Next, one can consider more specifically *what* information to provide. That information should address the "why" question, the goals and objectives, preferably from the point of view of the recipient of the information. What information is desired and useful, vs. what is possible? What information would be of interest and valuable? What information would strengthen confidence among parties, vs. what might be counterproductive, vs. what might be irrelevant? What types of information can be considered? Possibilities are endless: documents, declarations, measured data, images. It is also instructive to paint the possibilities on a spectrum of value. Typically the information will be anecdotal or suggestive, rather than complete and comprehensive.

When would the information be provided? The transparency could be a one-time event, or an ongoing promise. It could be offered at regular, predictable times, or only on occasion as desired by either of the parties. Information may be "pushed," i.e., sent out as decided by the supplier, or "pulled", i.e., retrieved if and when desired by the recipient. The provision could be prompt, or with some time delay.

What is the expected result of the information sharing? Is there any expectation of feedback, such as comment, questions, or even just acknowledgment? What is important to know after release? Was the information appreciated? Understood as intended? Valued? Looked at? Or does it suffice to consider "no news is good news"? Arguably, some benefit must be expected, or it would be nonsensical to expend the effort in the first place. A difficult technical question is how to measure or assess the results from transparency. If one cannot measure the benefit, however, then it is impossible to assess the cost/benefit for the transparency.

Unexpected results of information sharing must also be considered—at least to assess potential threats and the vulnerabilities that might be introduced through providing information. Collateral damage arising from the potential misuse of information, whether by the intended audiences, or leaks to unintended audiences, presents a risk.

How to supply the transparency is where the technical solution becomes especially important. Generally the desire for low-cost, low-impact solution involves some automation of the information sharing process. The process needs to employ a trusted mechanism that addresses security concerns. Could information be recalled if necessary? How would the system employ a review pipeline? Some level of oversight or audit will be necessary.

2.3. Considerations in "receiving" transparency

The recipient of transparently-offered information has a corresponding set of considerations. Typically, the situation is one of desiring to receive certain information and then finding a way to motivate another party to disclose it. But here we will instead assume that we are past that step: the information will be or has been provided. Knowing that another party is providing the information, the recipient at the very least needs to decide: What do we do with it?

The "why look at it?" consideration is relatively easy. The information is available; it might be useful, so take a look. Why not?

What to do with the information presents two possibilities: ignore it or act on it. In the latter case, *how* to deal with received information is the major consideration. Especially if this is an ongoing (vs one time) provision of information, is there a business process to manage it? Who is the responsible point of contact? What is the pipeline to others who "need to know", either the raw information as-is, or some derived result, such as an analysis or summary?

Several questions must be answered in the course of analysis and evaluation:

1) How well do I trust this information? Can I be sure that I know where it came from, that it hasn't been tampered with (integrity), that its attributes are valid (e.g., time stamps)?

2) How valuable is this information? Do I care about it?

3) Assessment: what does this information tell me? Is there anything else, besides the intended message? Does it raise any questions? Is it consistent with other sources of information? Is there anything missing?

4) Next steps: Is there any follow-up that needs to be done internally? What consequent action is appropriate? Is a reciprocal response expected, or just an acknowledgment?

2.4. Considerations for multilateral arrangements

Multilateral arrangements introduce significant complexities for transparency. Immediately there is now a greater likelihood of facing a diverse audience, which could limit the scope of information a state is willing to share.

One way to deal with the differing interests is through "compartmentalization": having separate groups within the multilateral group. But what are now the business rules governing the way information and communication takes place within the arrangement? They can quickly become complex.

Authentication now becomes more important, which is the technical implementation for positively identifying the source of received information. Otherwise it is possible in principle for one party to impersonate another. Authentication also helps to preserve the integrity of information, identifying what is truly a genuine version. Especially as more recipient parties are involved, it is more likely to have multiple copies of information in circulation.

"Trust" is a key concept for voluntary sharing arrangements. Generally, trust needs to be the *outcome* of the transparency; the underlying system should not assume a trusted arrangement in the first place.

Any transparency arrangement needs to be able to add new members, or deal with members departing, seamlessly. Such extensibility requirements have many technical implications. The architecture or topology is important—how are parties connected with another. Is there a centralized location where information is exchanged (hub and spoke); is it instead a maze of two-way connections

between each pair of participants; or are there other strategies?

2.5. Considerations for all parties to transparency

All parties to a transparency arrangement would have many common, overarching issues. The topics suggested here are not comprehensive, but provide just a starting point.

A critical concern is security. What are the threats? What are the risks from the information sharing? Information surety is a paramount concern for transparency arrangements, not just for safeguards.[2]

Another concern is resilience, especially in how to deal with abnormal or unanticipated situations, such as accidents or system failures. There must be alternate communication channels for resolving various questions and problems. The arrangement must also be able to survive periods where the environment—outside the sharing arrangement—is not necessarily "cooperative."

How long is shared information available or retained? It is conceivable that information may also be used for safeguards. In that case, might there be there any conflicts?

Metrics--tools that can indicate objectively the value of transparency—are important to establishing its importance and justify sustaining the investment. Without them, it would be difficult to assess the cost/benefit of transparency.

Particularly bilateral arrangements should consider regional expandability: How might additional parties join the cooperation? At the very least, an arrangement needs to consider the "outside" environment, paying attention to how the sharing arrangement may be perceived by other states. Does the transparency arrangement itself need to be transparent to outsiders?

In certain situations, there may be a role for an intermediary. Can two parties cooperate directly, or do they require a trusted third party to facilitate an arrangement?

3. Technical implementation

All of the foregoing discussion suggests that the entire undertaking of transparency implementation entails a great deal of work.

For an enduring solution, there are significant benefits to automating as much of the process as possible to reap cost efficiencies. If designed well, technical solutions to automate a transparency arrangement can do much more than simply collect, transmit and store data. Two aspects in particular are critical: (1) that the technical solution accurately implements the requirements of a viable sharing arrangement, and (2) that the solution reliably implements appropriate security for the intended sharing arrangement, which is key to promoting trust between the parties.

Comprehensive requirements for information sharing are essential to enable the design and development of a system solution. Any technical approach will need to identify and develop the technical building blocks for the information sharing, both the technologies (such as remote monitoring) and the procedural elements. The scope may involve research and development, training, prototype experiments, testing and other activities.

Implementation is concerned with what methods can be used to generate, authenticate, transmit, store, archive, access, protect and evaluate information. How are availability and reliability assured? How will the system be maintained? How to accommodate new technology developments or obsolescence? Personnel turnover? What approvals will be necessary? What testing will be required? The answers to all of these and other questions will comprise the functional requirements for an information-sharing network.

4. Discussion

4.1. Implications for safeguards

State to state and regional cooperation on nuclear nonproliferation might at first seem unnecessary, particularly to those who believe that the compliance-based safeguards system is fully sufficient. But the argument may be irrelevant, as various states are contemplating transparency measures anyway.[3] It is critical *how* they implement such information sharing, so that the likelihood of a benefit exceeds the possibility for negative impact. Although transparency could complement IAEA measures by addressing the kinds of questions the safeguards system is not designed to answer, there is (in principle) a risk that the two systems could give inconsistent messages.

4.2. Implications for regional systems

European safeguards under the Euratom treaty was a parallel development along with the IAEA system, yet has managed to evolve and continue under a partnership agreement with the IAEA. The Brazilian-Argentine Agency for Accounting and Control of Nuclear Materials (ABACC) was a special case that was instrumental in facilitating Argentina and Brazil's accession to the Treaty on the Nonproliferation of Nuclear Weapons (NPT); it too has managed to evolve and continue. There remains a possibility that other regional systems could appear, although it is not clear just what they would look like. Nevertheless, voluntary cooperation employing transparency is a distinct possibility. Nascent efforts at state-to-state cooperation in the area of nuclear nonproliferation could eventually mature into future regional systems. It is therefore timely that the technical details for implementation are given proper attention early in the development.

5. Summary

Voluntary information sharing ("transparency") between states is complicated, involving considerations from the point of view of the supplier state, the recipient state, and greater complexity when in a multilateral context. Many technical details are involved, and security measures are necessary to mitigate risks. Nevertheless, such nuclear nonproliferation cooperation can complement IAEA safeguards and strengthen the nonproliferation regime. A comprehensive, systematic approach is necessary to ensure successful implementation.

Approved for release:
SAND2011-3415C

Acknowledgments

The author thanks past and present colleagues at Sandia National Laboratories, as well as many international partners in the U.S. Department of Energy's international nuclear safeguards collaborations with partners in the Asia-Pacific, South America and Europe, particularly with the Japan Atomic Energy Agency and the Korea Institute of Nonproliferation and Control, who have contributed valuable discussions related to the technical implementation of transparency. The International Nuclear Safeguards and Engagement Program of the U.S. National Nuclear Security Administration has been a generous sponsor of ongoing work by Sandia National Laboratories in this area. Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

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When is a mine, a mine?

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Abstract:

Under the Additional Protocol states are required to inform the IAEA of the location, operational status and the estimated annual production capacity of uranium mines and concentration plants and thorium concentration plants. There are many possible combinations of mines, mills and processing plants involving various minerals that could finally produce uranium and/or thorium. Increasingly uranium is and will be produced from co- or multi-product mines. Phosphates and even coal ash are being seen as potential sources of uranium. While uranium and thorium may initially be considered part of tailings and/or waste in one phase of resource extraction, nuclear material may be the primary product in subsequent phases. The rise and fall in the price of uranium results in a corresponding movement in uranium exploration activity. Are all these activities meant to be captured under the Additional Protocol? The ultimate question is: when is a mine, a mine? There is little definitive guidance on this from the IAEA. This paper proposes a pragmatic approach to providing the IAEA the required information while being sufficiently transparent about uranium mining operations.

Keywords: uranium**;** mining; Additional Protocol;

03 NDA III – Measurements

Verification of the enrichment of fresh VVER-440 fuel assemblies at NPP Paks

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Abstract:

An NDA method was developed for verifying the ^{235}U enrichment of homogeneous and profiled unirradiated VVER-440 reactor fuel assemblies by gamma spectrometry, by which about 30 assemblies were tested. Five of them was homogeneous, with ^{235}U enrichment in the range 1,6 % to 3,6 %, and the others were profiled with pins 3,3 % to 4,4% enrichments. Two types of gamma detectors were used for the test measurements: 2 coaxial HPGe detectors and a miniature CdZnTe detector, which fits into the central tube of the assemblies. In this way it was possible to obtain information from both the inside and the outside of the assemblies.

It was shown that it is possible to distinguish different types of assemblies within a reasonable measurement time. The measurement time was 1000 sec for each assembly. For the HPGe measurements the assemblies had to be lifted out from their storage rack, while for the measurement with the CdZnTe detector the assemblies could be left at their storage position, as it was shown that the neighbour assemblies do not have an effect on the results of the measurements inside the assemblies' central tube. The measured results were compared with a simulation calculated by the MCNP method. A recommendation for the optimal approach to verify the ^{235}U enrichment of fresh VVER-440 reactor fuel assemblies is given.

Keywords: fresh fuel assemblies; U-235 enrichment; HRGS/MRGS

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Enrichment measurements

Necessity of determining enrichment of the fresh fuel was raised in Paks NPP. It is important from the viewpoints of IAEA and EURATOM and the safe work of the plant. It may happen that the shipped fresh fuel is not assembled of pins of the declared enrichment. No deliberate action is presumed, but such a case may occur due to the high number of assemblies.
If the deviation is revealed during reactor operation only, the shutup of the reactor, dismounting and refueling the core may cause high expenses and interruption/drop-out in the power production.
Preliminary experiments were carried out for determining enrichment of the fresh fuel assemblies (FFAs). Planar and coaxial Ge and a CZT detector were used. The enrichment was determined from the uranium gamma spectrum taken by the planar detector in the 90 – 120 keV region by using the MGAU code. Results were obtained in agreement with the nominal enrichments in the case of older type (not profiled) assemblies. Results obtained from the use of the MGAU code differ significantly from nominal values of the profiled assemblies, owing to the configuration of the latter, namely pins of lower enrichment are in the outer part, whereas those of higher enrichment are inside, see Figs 1, 2. Due to the self-absorption, the lower energy gamma-rays arrive at the detector from the outer pins of the assemblies, looking to the detector's direction, whereas the contribution of the gamma-rays emitted by the inner pins is smaller.

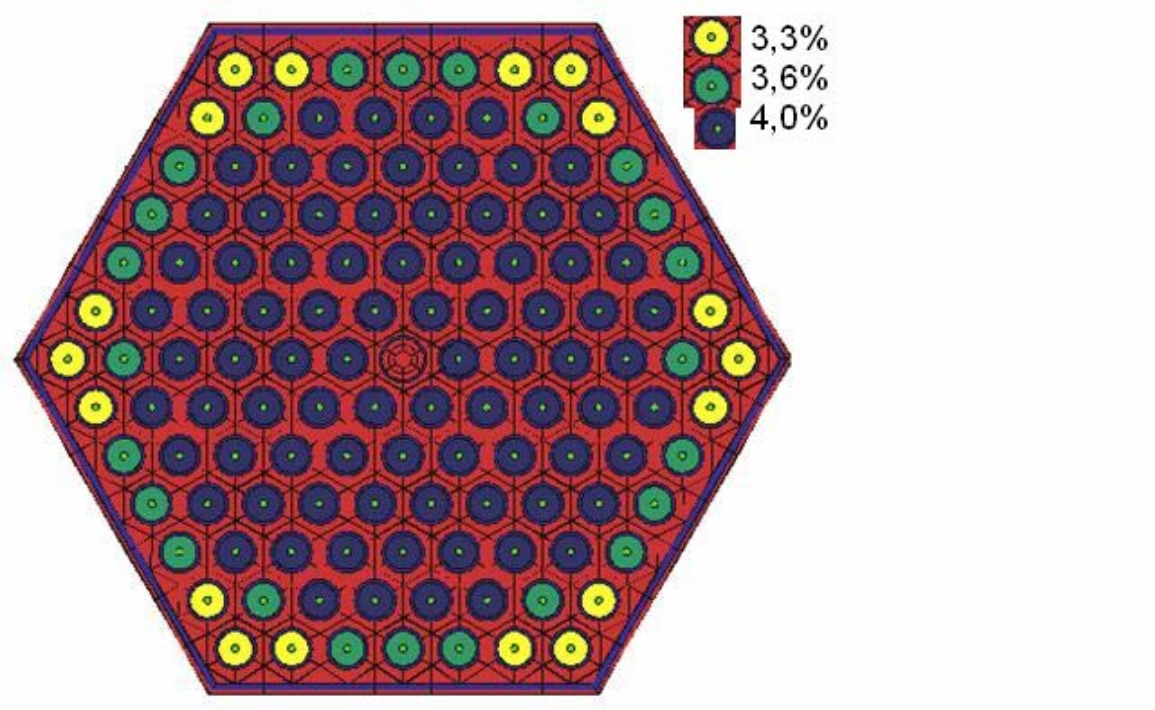

Fig. 1. Schematic diagram of the 3,8% profiled assembly

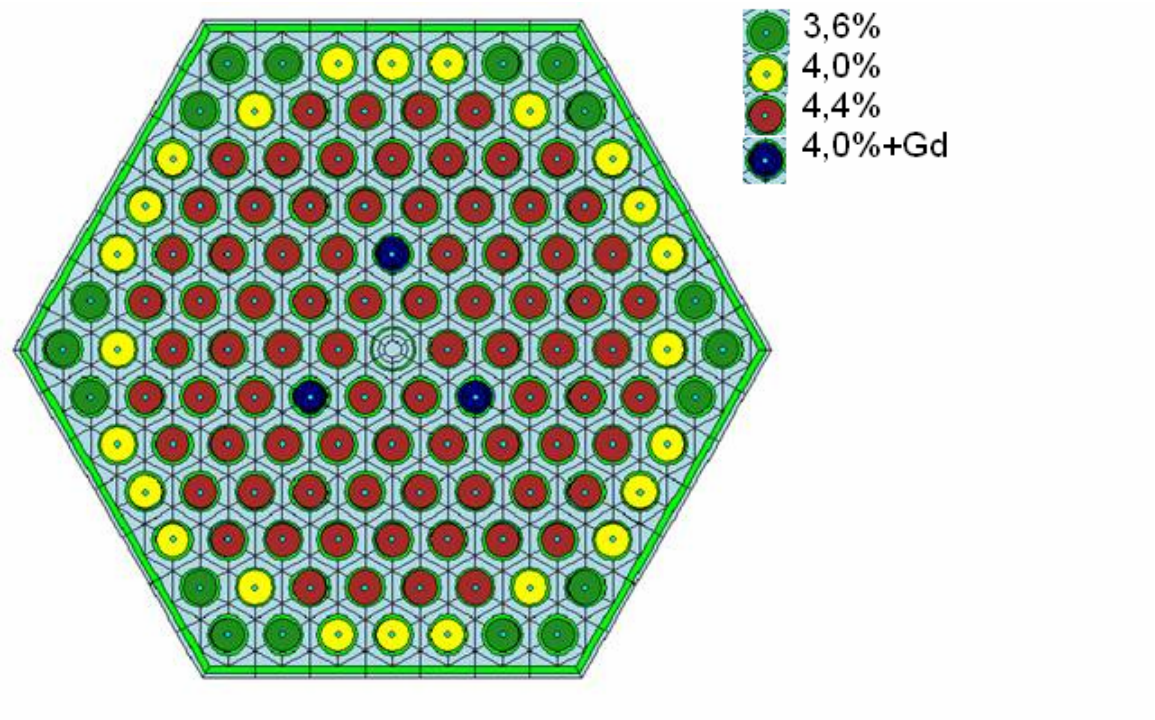

Fig. 2. Schematic diagram of the 4,2% profiled assembly

While measuring by the coaxial detector, the enrichment was determined on the basis of the intensity ratio of 186 keV (U-235) to 1001 keV (Pa-234m) peaks. The activity of the latter is in radioactive equilibrium with that of U-238 already 3 months after the enrichment. Absolute enrichment cannot be evaluated, however, because the ratio depends considerably on geometry. Also, the attenuation of the two peaks differ substantially, due to their energy differing very much. Therefore, the method can be used for enrichment determination after calibration by standard assemblies only, and by keeping a stable geometry. Owing to the higher penetrability of the 186 keV energy gamma–rays over those at about 100 keV, the method is capable to provide information on pins being inside the assemblies. Nevertheless, getting an „insight" into the assemblies is limited to a few rows' depth only. Insofar as the geometry can be kept stable very precisely and the amount of U-238 is nearly the same, it is practically enough to measure the 186 keV peak area instead of measuring peak area ratios 186 keV/ 1001 keV.
Measuring by the CZT detector completes the measurement from outside so far as it can be placed into the central tube of the assembly, and in this way the 186 keV gammas emitted by the inner pins can be detected. The size of the detector makes it possible to detect the 186 keV gammas well, but 1001 keV gammas are detected by a low efficiency already. Fortunately, the central tube defines the measuring geometry well.
Because of the self-absorption of uranium present in the assembly, the 186 keV gammas come to the detector from the pins being in a few circles around the central tube. The contribution to the peak area of pins situating farther outwards is negligible.

Enrichment verification of the assemblies goes as follows:

1. Spectra of assemblies of homogeneous composition are taken by the planar Ge detector and evaluated by the MGAU code. It is checked that the measured values comply with the declared ones (1.6, 2.4, 3.6 %; all the three, but two different are

necessary out of them at least). These assemblies will be the standard ones, by which a method is to be calibrated as follows:

2. The spectra of the assemblies assigned as standards are taken by the coaxial Ge detector being fixed on the inspection stand for size check. The background spectrum is taken as well, and it is to be repeated whenever the situation of the assemblies essentially changes in the stand. Spectra of the assemblies to be examined are taken, too. The position of the detector on the stand is to be assured stable at half-height of the assemblies, at a distance of about 30 cm, see Fig. 3. At such a distance the measurement uncertainty of the 186 keV peak area decreases below 0.5% during a measurement time 500 s.

Fig. 3. Detector stand and assembly in measuring position

3. Spectra of the standard assemblies and of those to be examined are taken by the CZT detector placed in the central tube at a defined depth, see Fig. 4. The necessary measurement time of the detector given is 2000 s for reaching an uncertainty of 1% of the peak area.

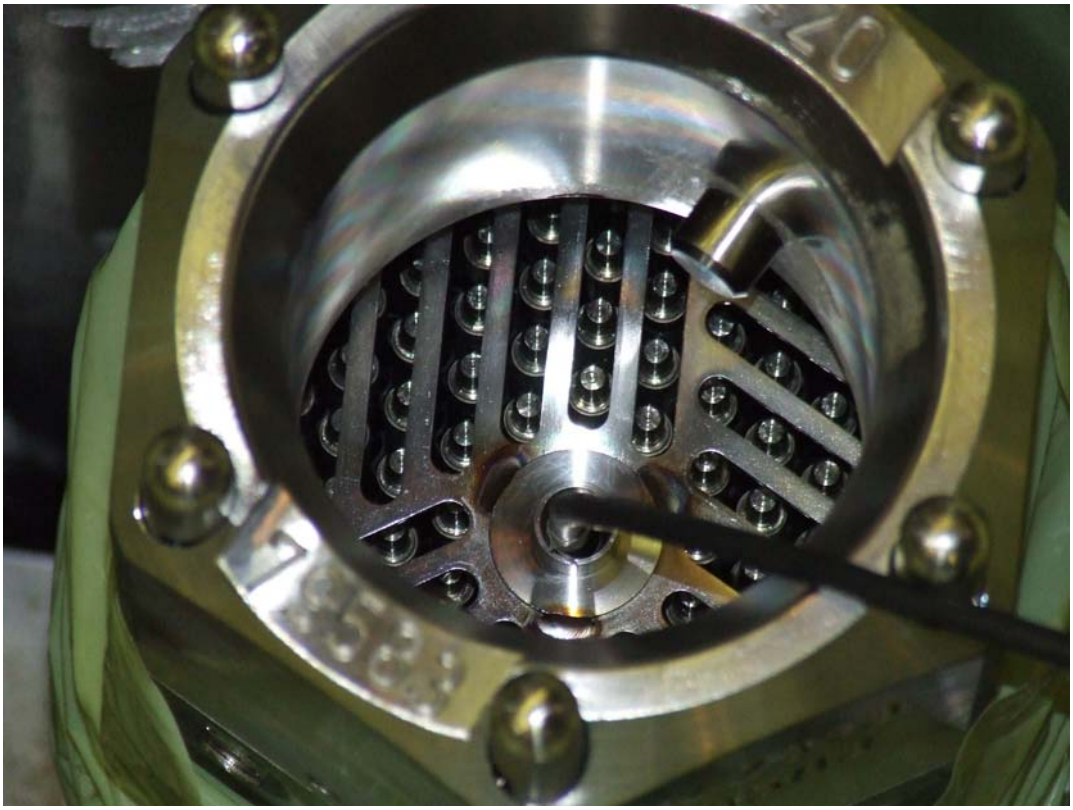

Fig. 4. CZT detector in the cenral hole of the assembly

Evaluation of the results

Count rate values of the 186 keV peak areas of the standard assemblies were plotted as a function of the enrichment. A straight line was fitted to the points, whereas determining the line parameters. The 186 keV count rates of the assemblies examined provide directly the average enrichment of „visible" pins. The calculations were to be performed for the measurements carried out by the Ge and the CZT detector.
These results, of course, would not agree with the declared average enrichment of the assemblies examined. The mesurement setup had to be modeled by the MCNP code see table 1.

Table 1. Calculated value of enrichment by MCNP method

Type of assemblie	Nominal Enrichment %	Calculated enrichment % from outside	Calculated enrichment % from inside
338	3.8	3.55	4.00
342	4.2	3.91	4.34

Results of the runs are to be compared to the enrichment values calculated from the measurements. The two data should agree within the measurement uncertainties.

Measurements carried out and results obtained so far

10% of 3 fresh fuel shipments arrived since having performed preliminary measurements were tested in the fresh fuel storages (FFS) 1 and 2 of Paks NPP. The equipment was calibrated 3 or 2 available standard assemblies of different enrichment in every experiment. Altogether 30 assemblies of 4.2% nominal enrichment, and also some ones of 3.8% nominal enrichment arrived earlier were checked. The assemblies were counted for 1000 s (Ge) and 2000 s (CZT). For determination of the repeatibility of the method all the six sides of the assemblies were measured by the Ge detector in some cases. The results verify the declared enrichment values.

Table 2. Examined assemblies of the first shipment FFS 1

Assembly	Nominal enrichment	Measured enrichment % Central hole, CZT detector	Uncertainty %	Measured enrichment % HpGe detector outside	Uncertainty %	Remarks
68787	1.6	1.59	0.1	1.61	0.1	standard
67791	2.4	2.41	0.1	2.39	0.1	standard
60537	3.6	3.60	0.1	3.60	0.1	standard
77769	3.8	3.98	0.1	3.60	0.1	earlier
77977	3.8	3.96	0.1	3.60	0.1	earlier
79255	4.2	4.34	0.1	3.95	0.1	
79264	4.2	4.32	0.1	3.92	0.1	
79241	4.2	4.25	0.1	3.90	0.1	
79235	4.2	4.40	0.1	3.95	0.1	
79271	4.2	4.24	0.1	3.92	0.1	
79260	4.2	4.31	0.1	3.96	0.1	
79286	4.2	4.41	0.1	3.95	0.1	
79249	4.2	4.37	0.1	3.91	0.1	
Average for 4.2%		4.33		3.93		

Table 3. Examined assemblies of the second shipment FFS 2

Assembly	Nominal enrichment	Measured enrichment % CZT detector, Central hole	Uncertainty %	Measured enrichment % HpGe detector outside	Uncertainty %	Remarks
67854	2.4	2.41	0.1	2.40	0.1	standard
60520	3.6	3.61	0.1	3.60	0.1	standard
79559	4.2	4.37	0.1	3.91	0.1	
79560	4.2	4.32	0.1	3.91	0.1	
79581	4.2	4.35	0.1	3.90	0.1	
79582	4.2	4.33	0.1	3.90	0.1	
79583	4.2	4.28	0.1	3.90	0.1	
79589	4.2	4.34	0.1	3.92	0.1	
79590	4.2	4.38	0.1	3.89	0.1	
79591	4.2	4.36	0.1	3.95	0.1	
79592	4.2	4.30	0.1	3.92	0.1	
79607	4.2	4.36	0.1	3.93	0.1	
Average for 4.2%		4.34		3.91		

Table 4. Examined assemblies of the third shipment FFS 2

Assembly	Nominal enrichment	Measured enrichment % CZT detector, Central hole	Uncertainty %	Measured enrichment % HpGe detector outside	Uncertainty %	Remarks
67854	2.4	2.4	0.1	2.40	0.1	standard
60520	3.6	3.6	0.1	3.60	0.1	standard
77791	3.8	n.a.	0.1	3.60	0.1	earlier
79527	4.2	n.a.	0.1	3.89	0.1	
79535	4.2	4.33	0.1	3.89	0.1	
79544	4.2	4.32	0.1	3.91	0.1	
79548	4.2	4.34	0.1	3.90	0.1	
79631	4.2	4.26	0.1	3.89	0.1	
79643	4.2	n.a.	0.1	3.92	0.1	
79674	4.2	4.28	0.1	3.87	0.1	
79695	4.2	4.30	0.1	3.89	0.1	
79707	4.2	4.33	0.1	3.88	0.1	
Average for 4.2%		4.31		3.89		

Further plans

Application limits of the method will be determined by MCNP simulations, knowing the measurement uncertainties. It will be examined whether it can be demonstrated that the enrichment of one or more pin in the assembly departs from the declared value. Moreover, in case of assemblies containing also pins doped with gadolynium that the direction of such a pin can be detect.
Assemblies as much as possible are to be examined by decreasing the measurement time, while keeping uncertainties as low as possible.
Due to the small size of the CZT detector, the measurement time cannot be reduced substantially, because of accuracy expectancies. Our aim is to design and purchase a

detector which, keeping a good resolution and small size, results in a shorter measurement time and/or lower uncertainty.

Modular Spent Fuel Attribute Tester Application

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Modular SFAT system was developed in the frame of the Hungarian support programme to IAEA safeguards. The detector house is watertight, 50mm lead shielding inside for side gammas. The built-in medium resolution detector is made of CdZnTe hemispherical, 500 mm^3 volume, connected to a mini MCA and controlled by a laptop. Each collimator is a 1 m long, ∅55 mm air filled stainless steel tubes, available up to 8 pieces and the neighbour effects are minimized by the collimator design. The bottom of the lowest collimator tube was taken into the head part of VVER-440 fuel assembly, so it's not needed to move fuel/object. The positioning stand is mounted on the railing of the refuelling machine. The whole system is possible to install and handle by two people.

A wide range of fuel assemblies and other objects was verified with the device. The measured fuel elements produced 662 keV peaks of ^{137}Cs and 1173 keV, 1332keV peaks of ^{60}Co, with good statistics, and 796 keV peaks of ^{134}Cs is appreciable in most cases. The 662 keV peaks identified even in extremely low burnup (order of GWd/tU) fuel with 6-7 y cooling time. This is a method for revealing undeclared irradiation in the verification of ^{60}Co sources. Tanks without fuel (construction part of damaged fuel assemblies) and canisters with repackaged damaged fuel (type T28 for parts of fuel assemblies and T29 with baskets for pellets only) were also tested.
The SFAT may give a good alternative in cases, when Cherenkov device is not usable, e. g. bad quality water, extreme long cooling time (up to 14 y), or low burnup.

Keywords: spent fuel assemblies; NDA method; MRGS

Introduction
The most widespread method for verifying the presence of the fissile material is the viewing of Cherenkov light. However it cannot be applied if the water is not clean enough and it doesn't identify the source of radiation. A more sophisticated method of verification is the SFAT where a medium resolution gamma detector identifies the source of radiation through a collimator tube. Such a device was designed and built in our Institute and it was tested in a series of verification problems.

Setting up of the device
Our apparatus consists of a detector house, which provides place for a 500 mm3 CZT detector and a lead collimator shielding the detector from side directions and a set of closed end (watertight) steel collimator tubes of 1 m length and 50 mm diameter, containing air (Fig. 1). The collimation itself is provided by the surrounding water.

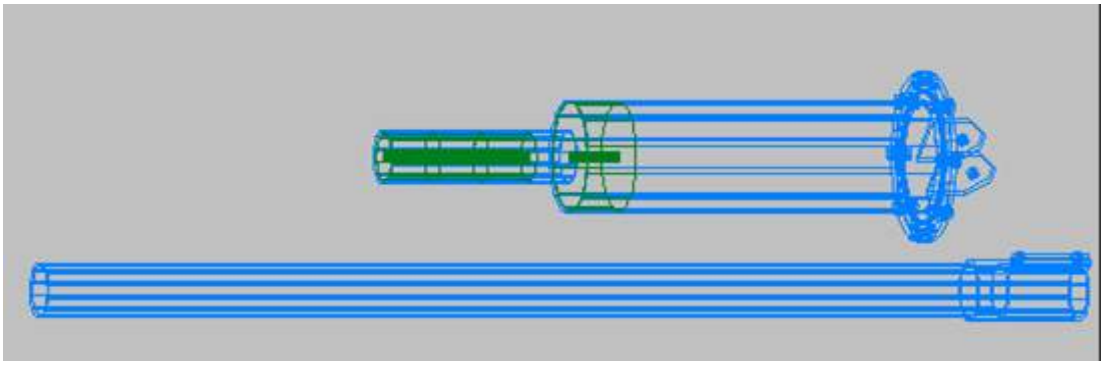

Fig. 1. Detector house and collimator tubes. The thick green line shows the volume inside the lead collimator.

The number of the tubes applied depends on the task. The maximum number of 6-8 tubes can be applied when the assemblies are stored in two levels and the assemblies in the lower level have to be verified. In most cases use of 2-4 collimator tubes is suitable while for assemblies cooled for a long time, use of 1 or 2 tubes provides good results. A part of the system is a stand for holding and positioning the detector and collimator tube system, which can be installed on the railings of the refuelling machine, or on a service bridge (Fig. 2). By the aid of the stand the mounted system can be moved above the assembly to be examined and sunk under water by the winch. A rough positioning can be done by moving the refuelling machine or substituting bridge, and by shifting the stand perpendicularly to the railing by the aid of rollers. Fine positioning goes by screws on the stand in two directions. The system can be mounted up to 4 collimator tubes laid just on the platform in advance and can be let down as a whole to the pond, while in the case of a longer collimator the individual tubes can be joined step by step and let the mounted part moving down. Detector house is connected at last.

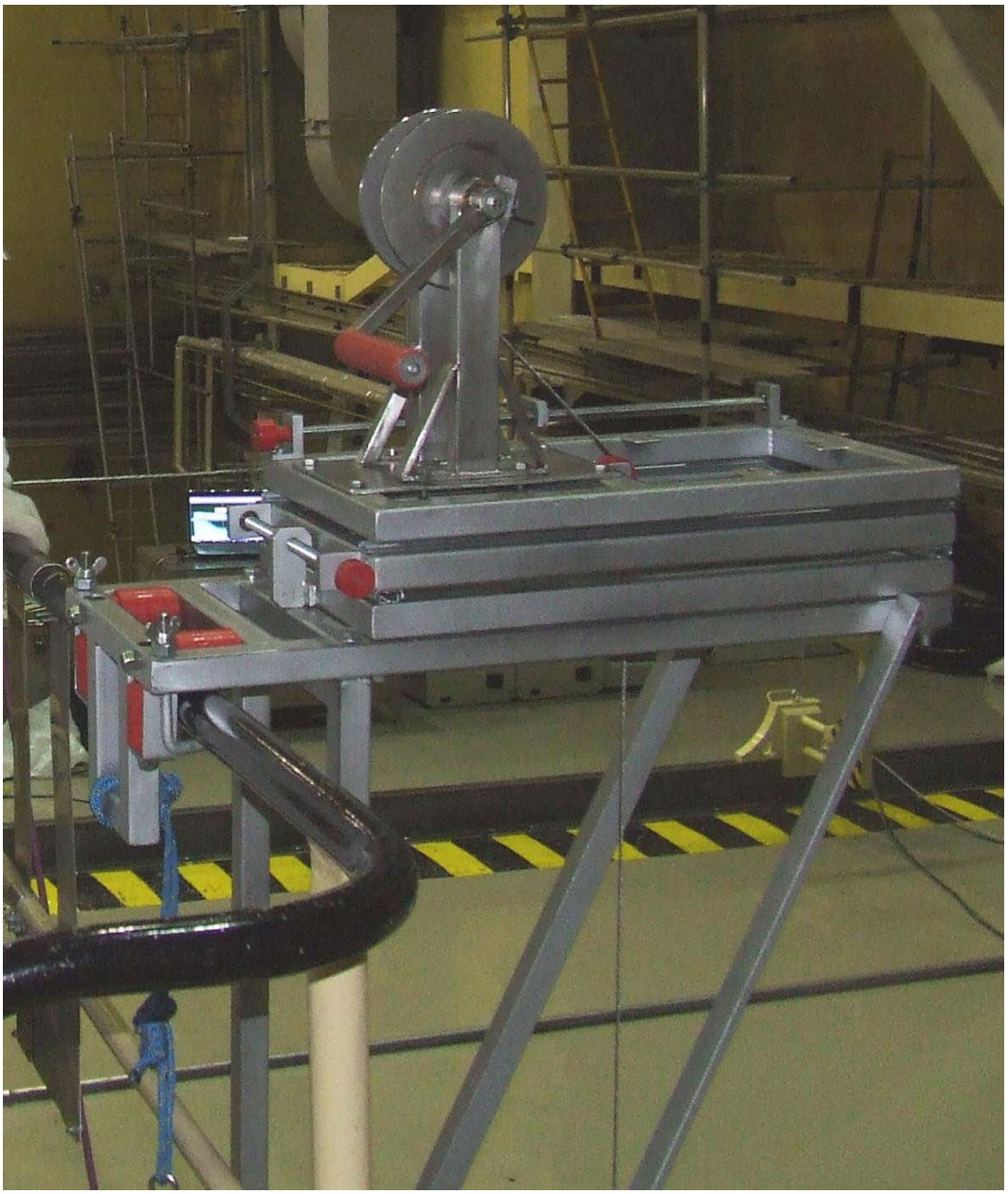

Fig. 2. The stand with the winch

For performing verification, the lower end of the collimator tube enters into the headpiece of an assembly until it neither impacts on the upper grid nor leans against the interior of assembly head, ensuring that the spent fuel and the detector are in line of sight through the collimator tube (Fig. 3). Positioning is supported by a TV camera and monitor as a surveying system. A camera mounted to the detector house may promote a more compact design.

Fig. 3. End of the collimator tube above the fuel assembly

Although it was developed for verifying VVER-440 assemblies, and also the positioning stand was designed to the refuelling machine established in the Paks NPP, the system can be used, with a minor change and transformation of the stand, in other plants as well.
The signal from the detector comes to the mini MCA analyser through a watertight insulated cable controlled by a laptop computer.

Applications
Assemblies and other objects stored in the spent fuel pond were examined by the SFAT. The spent fuel assemblies produce a 662 keV peak from the fission product Cs-137 as well as 1173 and 1332 keV peaks from Co-60, an activation product originating from the headpiece of the assemblies. If the cooling time of the assembly is less then 6 years, the 794 keV peak of the Cs-134 (2 y half life) can be observed as well. However, for very fresh assemblies (CT<1 y) the Compton-tail of the peaks of Zr-95 (half-life 64 d) and other short-lived activation products can cover the Cs peaks (Fig. 4).

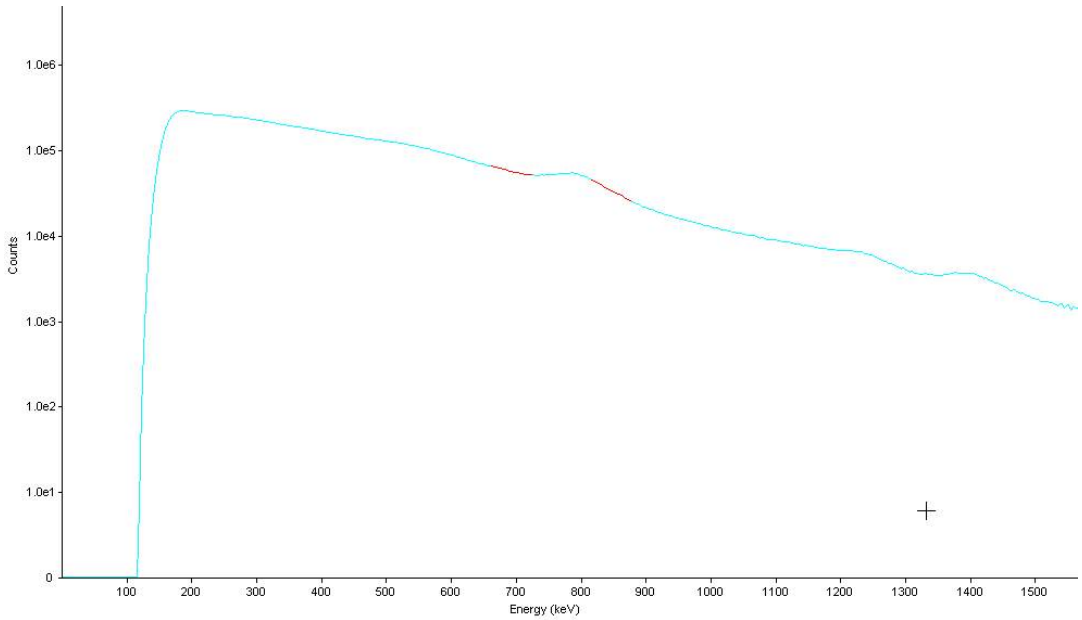

Fig. 4. Spectrum from the spent fuel assembly with CT= 8 months

With medium cooling times both Cs-134 and Cs-137 (Figs. 5-6) peaks can be clearly observed. As the intensity of the Cs-134 peaks is proportional to the square of the burnup, so the burnup can be assessed from the extrapolated initial value of the intensity.

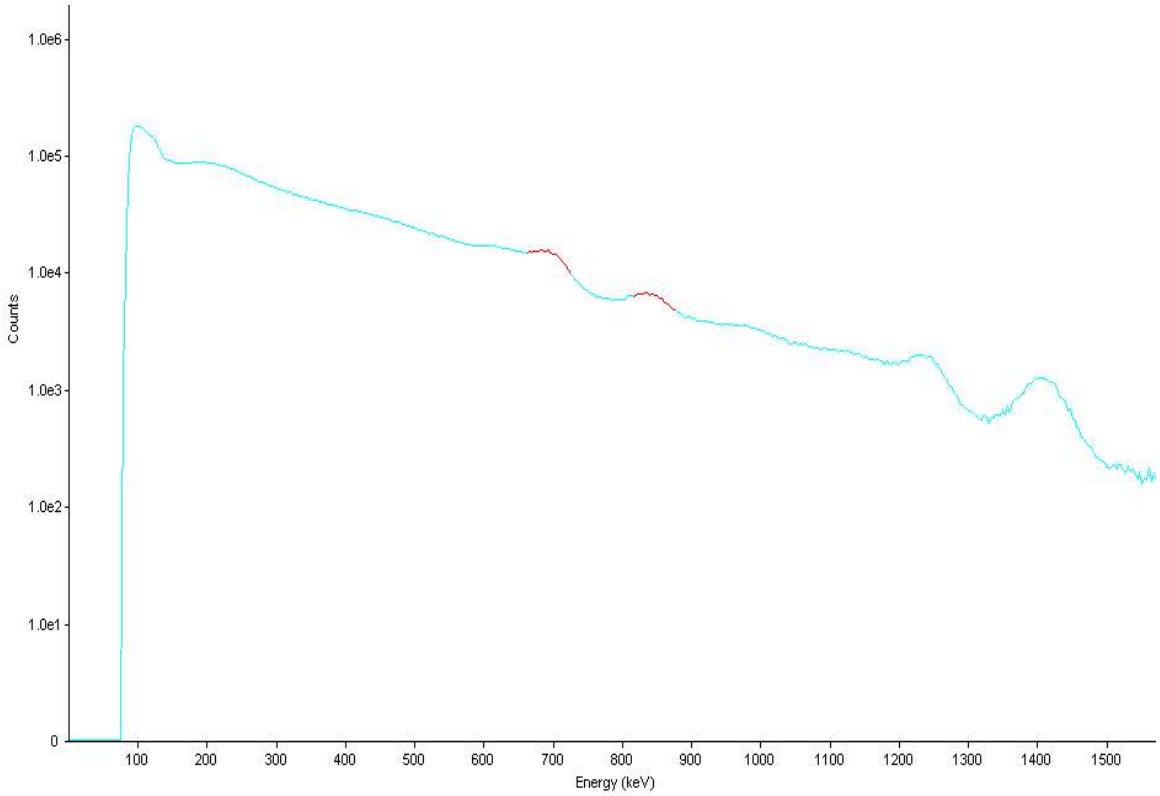

Fig. 5. Spectrum from the spent fuel assembly with CT=2.7 y, BU=22.6 GWd/tU

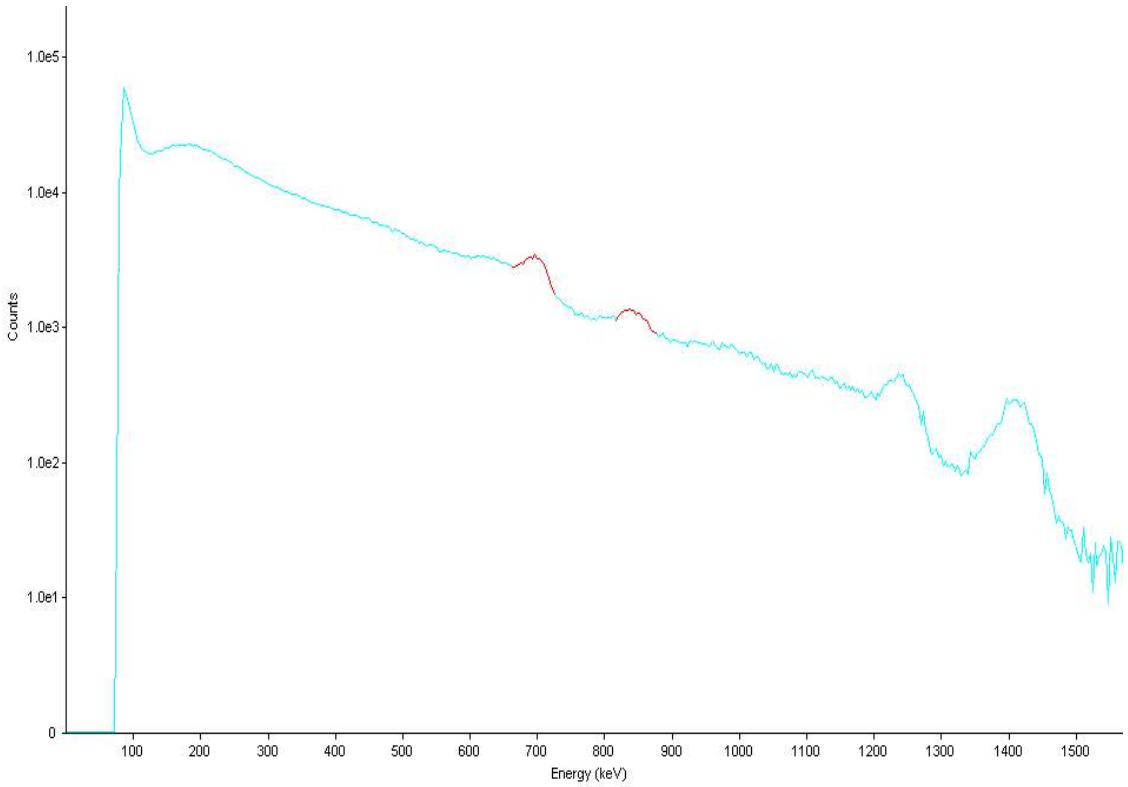

Fig. 6. Spectrum from the spent fuel assembly with CT=3.7 y, BU=41 GWd/tU

Due to the 30 y half-life of Cs-137, the 662 keV peak can even be revealed after an extreme long ((>14 y) cooling time (Fig. 7) or very low burnup (order of a few GWd/tU) up to 6-7 y cooling time, when the Cherenkov viewing device (ICVD) is not suitable for verification. The measuring time needed for a relyable spectrum lies between 300 and 1200.

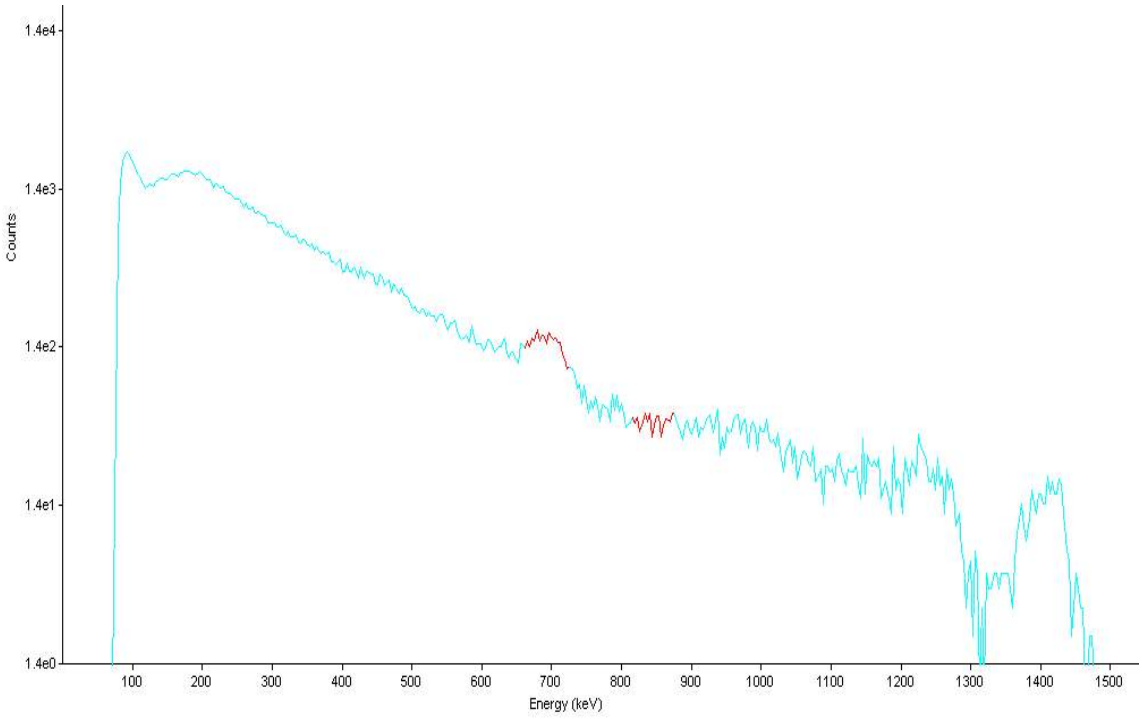

Fig. 7. Spectrum from the spent fuel assembly with CT >14 y

With the same method, we verified the fissile material content of those canisters which contained damaged spent fuel originating from the 2004 incident, and demonstrated the absence of the fissile material in canisters containing only the head and foot parts of the assemblies. In addition we demonstrated the absence of fissile material in the containers of irradiated Co-60 sources.

Summary

SFAT is useful, whereas acknowledging its limits, for verifying fissile material in fuel assemblies, containers and other objects. Detection of undeclared irradiation is also possible, even in the presence of Co-60. Necessary measurement time is 300-1200 s pro assembly/object, depending on its parameters.

Even if it cannot be a rival of ICVD in comfortable employment and easy evaluation, in cases where the water in the pond is of too high level or bad quality, burnup is very low or cooling time is too long, as well as if the task is to verify an assembly in the lower rack, it may be a good alternative for substituting ICVD.

Determining the mass of nuclear and radionuclide samples by radiometric calorimetry

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Abstract

This paper reports on results of measurements and analysis carried in order to recalibrate and assess the performance of our newly upgraded Small Sample Calorimeter (SSCAL) in the Performance Laboratory of the JRC Nuclear security Unit in Ispra (Italy), using calibrated electric samples and standard reference nuclear materials. The SSCAL is a heat flow calorimeter, which works by measuring the voltage generated by heat-emitting sample across a thermal gap based on a thermopile cup technology. Preliminary results of calorimetry measurements carried out on reference PuGa samples and on well characterized ^{241}Am samples will be presented.

Keywords: Calorimetry, NDA, PuGa, ^{241}Am

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1. Introduction

Calorimetry remains one of the most accurate non-destructive assay (NDA) technique for materials containing plutonium, when combined with accurate isotopic analysis using high resolution gamma-ray spectrometry. This made it the primary measurement method in the USA during many decades from as early as 1940's when it became arguably the most important part of plutonium accountability. For, the technique is accurate, unbiased and unaffected by geometry and sample matrix effects due to the fact that the magnitude of the heat flux leaving the sample container at equilibrium is not affected by the matrix. Furthermore calorimetry requires no physical standards to represent the samples of interest.

As previously described and reported [1,2] the SSCAL has since been extensively tested and its performance evaluated as the first of a new generation of plutonium calorimeters based on thermopile technology. It was subsequently used for instance at the Institute of Isotopes of the Hungarian Academy of Science in Budapest (KFKI) for the characterisation [3], as dictated by IAEA safeguards requirements, of hundreds of PuBe and AmBe sources that came into the country during the soviet union era mainly. Most recently, seven ^{241}Am sources produced by ITU to be used cross section measurements by the JRC Institute for Reference Materials and Measurements (IRMM in Geel Belgium) have been measured using calorimetry in conjunction to gamma and neutron spectrometry where needed. These measurements were compared to declared masses measured at ITU using various means [4].

2. Description and general considerations

The model 601 transportable small sample calorimeter (figure 1), measures heat producing samples with thermal powers in the range 0 - 200 mW. The SSCAL, developed by ANTECH (Oxfordshire, UK) is one of a new generation of calorimeters that employ thermopile technology in conjunction with traditional nickel resistance thermometry. Thermopiles consist of a serially interconnected array of thermocouples, which produces an offset free Seebeck voltage, proportional to the temperature difference between two sites. The SSCAL is a heat flow calorimeter, which works by measuring the voltage generated across a thermal gap, consisting of a thermopile cup, when it contains a heat-emitting sample. The SSCAL has a dual measurement chamber system of well size 50 mm diameter and 100 mm high.

Figure 1: Photo of the SSCAL

Thermal equilibrium of the calorimeter is achieved by maintaining a zero heat flow between the heat-sink and the cylinder surrounding it. If the heat-sink is warmer than the outer cylinder, as is the case when a sample is present, heat will flow outwards from the heat-sink, towards the outer cylinder thus generating a voltage in the annulus thermopile. The voltage generated in the thermopile cups is measured by a dedicated two channel Keithley nanovoltmeter, wired directly to the cups. All other measurements are performed by a Keithley source meter via a scanner. Heat flow calorimeters are thus in their nature devices used to extract the mass of radioactive materials by measuring their heat output, and with the increased emphasis on safeguarding of nuclear materials, radiometric calorimetry had become one of the most preferred and powerful non-destructive (NDA) measurement techniques for plutonium and tritium which provides a reasonably high degree of accuracy and precision. However calorimetry often requires long measurement times, which may be a major disadvantage.

This has become apparent in measurement and testing campaigns carried out on tens of radioactive sources at the KFKI institute in Budapest [3] and here in Ispra, using our small sample calorimeter (SSCal). Upgrading our SSCal, some details of which are given below has thus become essential as one seeks to reduce measurement times and improve the accuracy, stability and reliability of the hardware and software systems.

In essence and amongst other changes, the outer insulation of thermal element was replaced with new and more performing insulation blocks, the plug units were redesigned, in order to improve thermal insulation (extended polystyrene), achieve greater isolation from ambient changes, and thus improve accuracy and also reduce the rate of heat leak to the ambient. A pre-heater system was provided which should result in a reduction of the measurement time by reducing the time taken for the sample to equilibrate to the measurement chamber. Further work is under way to assess corresponding effects and improvements.

3. Calibration of the SSCAL

As described above the SSCal is provided with two electric calibration samples which supply a calibrated power to one cup by means of a Power Amplifier, controlled by a DAC (digital to analog card) mounted on the computer and measured by the source meter via a resistor plate and a multi-channel scanner.

On the return of the SSCAL to Ispra and over many weeks, about 100 electric sample measurements were carried in the A-B mode with the power applied to cup-A in the range from 0.01 to 200 mW. Three sets of calibration parameters were subsequently produced for there different range of powers of interest: 0-5 mW, 0-20mW and 0-200 mW which will improve precision and accuracy for given sample output. A linear fit was applied which relates the Voltage measured by a calibrated Keithley nano-voltmeters to the applied heat power as follows:

$$\text{Applied Power(mW)} = \boldsymbol{a} + \boldsymbol{b} \times \text{Measured Heat Flow (mV)}$$

Table 1 gives the calibration parameters and plots to be used for the SSCal measurement in the CupA-CupB (differential) mode when power is applied to Cup A. The stared 0-200 mW calibration parameters correspond to a fit performed with all 10 points repeated measurements for 0.1, 1 and 5 mW averaged and inserted as one point (not 10) to avoid excessive biasing to lower values of heat flows.

range	Offset (*a*)	slope (*b*)
0-5 mW	-0.01206	8.33632
0-20 mW	-0.0212	8.37836
0-200 mW	-0.02818	8.39332
0-200 mW *	-0.02998	8.3947

Table 1: Cup A – Cup B Calibration coefficients

4. Measurement of Nuclear Samples

Subsequent to the above electric sample calibrations, the performance of small sample calorimeter was assessed by measuring a set of well-characterised reference plutonium standards, provided and certified by AEA Harwell UK. Two groups of PuGa sources were used in this work. One, labelled D00185 (sources 201 to 211) containing 21% of ^{240}Pu and the second labelled N.73240 (sources 15 to 37) containing 6 % of ^{240}Pu.

4.1 PuGa samples (21% in ^{240}Pu)

This group (D00185) consisted of a set of double encapsulated plutonium gallium (plutonium content 96.409%) alloy samples whose isotopic compositions (in weight %) of the samples, as on 10 May1996, are given in Table 3 below together with that of ^{241}Am which in terms of ^{241}Am to Pu weight ratio amounts to 0.0186. The Pu discs (0.6 mm thick) are made in the form of Pu-1.5% Ga alloy sandwiched in 46 mm diameter stainless steel capsules of total thickness about 1.5 mm and wall thickness 0.5 mm. Results of the Calorimetry measurements carried out on the PuGa samples with the SSCAL operated both within and outside the climatic chamber are given in Table 4. Where as for most samples, measurements were repeated more than once (13 times in the case of PuGa206 and about 2-3 times for all others), the average values are given. The build up from beta decay of ^{241}Pu and decay of ^{241}Am is taken into account and the specific heat taking into account all isotopes is 6.941 mW/g of Pu.

Isotope	Iso.compo wt %	rsd %	Specific Power mW/g (error)	Half Life (y)
Pu-238	0.1336	0.04	567.57 (0.26)	87.74
Pu-239	75.6606	0.03	1.9288 (0.0003)	24119
Pu-240	21.4898	0.07	7.0824 (0.002	6564
Pu-241	1.9510	0.93	3.412 (0.002	14.348
Pu-242	0.7651	0.38	0.1159 (0.0003	376300
Am-241	1.86	0.02	114.2 (0.42)	433.6

Table 3: Isotopic composition, specific power and half lives of the D00185 plutonium standards

Samples	Declared Pu Mass (g)	Decay corrected Pu Mass	Meas Heat Flow mW	Measured Pu Mass (g)	1-mea/dec
PuGa202	0.0175	0.0174	0.0860	0.0120	30.92%
PuGa203	0.0447	0.0443	0.2967	0.0430	2.86%
PuGa204	0.0938	0.0928	0.6424	0.0927	0.13%
PuGa205	0.1804	0.1786	1.2384	0.1790	-0.23%
PuGa206	0.4394	0.4349	3.0069	0.4348	0.04%
PuGa207	0.8838	0.8746	6.1247	0.8847	-1.15%
PuGa209	1.8716	1.8522	12.8098	1.8503	0.10%

Table 4 : Results of calorimetry measurements of PuGa samples (D00185) using low Range calibration parameters (see text for further explanations).

4.2 PuGa samples (6% in ^{240}Pu)

This group (N73240) of certified samples consisted of a set of double encapsulated plutonium gallium (plutonium content 97.935%) alloy samples whose isotopic compositions (in weight %) of the samples, as on April 1996, are given in Table 5 below together with that of ^{241}Am which in terms of ^{241}Am to Pu weight ratio amounts to 0.00126 (ref. July1996). Finally, the Pu discs (0.2 to 0.6 mm thick and 2 to 36.5 mm diameter) are made in the form of Pu-1.5% Ga alloy sandwiched in stainless steel capsules. Results of the calorimetry measurements carried out on these PuGa samples with the SSCAL operated within the controlled environment of the climatic chamber are given in Table 6. These samples have the advantage in that the amount of Am contained and thus the corresponding heat effect uncertainty in the determination of the Pu content is minimised. The effective specific power equal to 114.2 mW per gram of Am is used.

Isotope	Iso.compo wt %	rsd %	Specific Power mW/g (error)	Half Life (y)
Pu-238	0.0117	0.01	567.57 (0.26)	87.74
Pu-239	93.8773	0.01	1.9288 (0.0003)	24119
Pu-240	5.9009	0.11	7.0824 (0.002	6564
Pu-241	0.1785	0.95	3.412 (0.002	14.348
Pu-242	0.0316	5.5	0.1159 (0.0003	376300
Am-241	0.126	0.001	114.2 (0.42)	433.6

Table 5: Isotopic composition, specific power and half lives of the 73240 plutonium standards.

Samples	Uncor. Ref Pu Mass (g)	Uncert.	Decay Corr. Pu Mass (g)	Measured Volage (mV)	Measured Heat Flow (mW)	Measured Pu Mass (g)	1-meas/decl
73240-32	9.40427	0.00623	9.391213	2.835001	23.7691	9.38	0.14%
73240-31	4.92842	0.00336	4.92158	1.485654	12.4416	4.91	0.26%
73240-30	1.94621	0.00134	1.94351	0.58767	4.9033	1.93	0.46%
73240-26	0.93969	0.00070	0.93838	0.288489	2.3918	0.94	-0.56%
73240-18	0.47163	0.00037	0.47098	0.146303	1.1982	0.473	-0.37%
73240-16	0.10302	0.00014	0.102876	0.031023	0.2466	0.097	5.44%

Table 6: Results of Calorimetry measurements of the 73240 PuGa samples

The mass of plutonium measured in these PuGa sources generally agrees with the declared mass within about 0.5% at 10 mW heat powers. The PuGa source number 73240-32 was found by Thornton et al. [1] to contain 53 ppm of Be (AmBe(α,n)) which is a source of random neutrons, based on

neutron coincidence measurements indicated by the Reals to Totals ratio they plotted against the totals rates. The effective specific power equal to 114.2 mW per gram of Am is used and 2.5346 mW/g for Pu .

4.3 ^{241}Am Source measurements

4.3.1 Calorimetry Measurements on the Am sources

Seven Am sources produced by ITU [4] to be used for cross section measurements by the JRC-IRMM have been sent to JRC-Ispra to be measured using the calorimetry in conjunction to gamma and neutron spectrometry, where needed. The preliminary results of our calorimetry measurements are shown in Table 7 and figure 3 in comparison to the declared masses as measured at ITU [4]. The effective specific power equal to 114.2 mW per gram of Am is used. Good agreement (on average within 0.5%) was obtained between the declared and the measured Am masses. Each source was measured with the SSCAL within the climatic chamber (ambient temperature about 22.5 degrees and humidity 45%) for about 300 minutes following an overnight stabilisation (with no source in cups) to minimise any residual effects and thus improve accuracy.

Neutron and gamma measurements have been performed on these samples at the Perla laboratories in Ispra and at the IRMM (gamma only) [6] in order to investigate the presence of impurities and any other competing gamma or neutron producing reactions. This work is in progress after which an extensive paper will be published in the open literature. All samples were measured or/and re-measured individually as well as bunched together to provide varying amounts of heat flows or masses. Coincidence neutron measurements carried in Perla Laboratories show that about 0.42 +/- 0.08 are registered as double counts against 0.03 for a random AmLi source.

	Sample	Declared Am Mass	Heat Flow	Measured Mass	Ratio
		Am (g)	mW	(g)	Mea/Dec
1	IRMM-4-run1	0.04098	4.70030	0.0412	-0.44%
2	IRMM-4 run2	0.04098	4.65333	0.0407	0.57%
3	IRMM-4 run3	0.04098	4.65150	0.0407	0.61%
4	IRMM-4 run4	0.04098	4.63029	0.0405	1.06%
5	IRMM-5	0.04121	4.68372	0.0410	0.48%
6	IRMM-6	0.0421	4.78016	0.0419	0.58%
7	IRMM-7	0.04184	4.75128	0.0416	0.56%
8	IRMM-8	0.04146	4.70117	0.0412	0.71%
9	IRMM-8run2	0.04146	4.70243	0.0412	0.68%
10	IRMM-8run3	0.04146	4.70365	0.0412	0.66%
11	IRMM-9	0.04209	4.77319	0.0418	0.70%
12	IRMM-4+5	0.08219	9.35802	0.0819	0.30%
13	IRMM-6to9	0.16749	18.98231	0.1662	0.76%

Table 7: Results of Calorimetry measurements on the IRMM ^{241}Am samples.

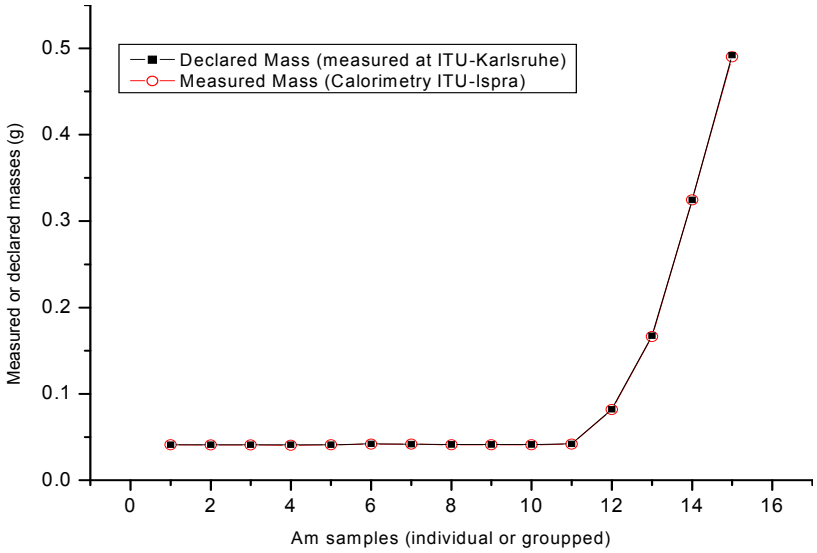

Figure 3: Mass of IRMM Am samples measured by the SSCAL calorimeter compared to the declared masses as measured at ITU.

5. Conclusions

The specifications required a precision of 0.2% or better at a thermal power of 10 mW. This was achieved in the differential mode usage of the SSCAL. Using plutonium standards and operating the calorimeter in the differential mode, a precision of 0.25% was obtained for samples of equivalent power about 1.0 mW. However, it is generally felt that a very high accuracy measurements can only be achieved if at least 3 hours measurements were performed. The calorimeter has been fully calibrated and successfully used to measure reference PuGa samples belonging to JRC-Ispra and Am sources of IRMM. The ^{241}Am sources were produced and their masses measured by ITU for IRMM who would use them for cross-section measurements. Our calorimetry measurements of the Am masses agreed extremely well with ITU declared masses. Further work to assess the calorimeter, including the use of a pre-heater to reduce measurement time, uncertainty analysis, neutron and gamma spectrometry measurements and analysis concerning ^{241}Am sources , etc.. is in progress and will subsequently be published.

Acknowledgment: Discussions with P. Peerani, B. Pedersen and J. Bagi during the preparation of this work is acknowledged and the assistance of Frison Santino with the handling of the sources is well appreciated.

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Design and Performance of the Digital Upgrade of the Mini Multi-Channel Analyser (DMCA)

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Abstract:

The Mini Multi Channel Analyser (MMCA) is a portable electronics module used for neutron and gamma ray Non-Destructive Assay (NDA) of nuclear material and is therefore an important tool in nuclear safeguards to perform certain classes of verification activities. To make use of advances in digital technology, communication protocols and standardisation, display technology, and user interaction paradigms, a digital upgrade of the MMCA is being developed in the frame of the German Support Programme to the International Atomic Energy Agency with cooperation of the Commission of the European Union.

The Digital Miniature Multi Channel Analyzer (DMCA) preserves the main functionality and compatibility with the existing MMCA. The DMCA is dubbed MCA-527 and is designed as a portable tool used for in-field inspection purposes where versatility, flexibility, reliability and ease of use are given a high priority. The transition to fast digitization of analogue signals and the integrated programmable digital signal processor enable increased performance and functionality in safeguards applications. As an example, improvements in operation of various types of detectors and acquisition modes are expected with the DMCA by virtue of digital filtering techniques. This paper presents the development status and hardware specifications of the MCA-527 and some performance results from measurements close to application.

Keywords: multi channel analyser; non-destructive assay; gamma spectroscopy; neutron detection

1. Description of design goals, framework

The Mini-Multichannel Analyser (Mini-MCA) or MCA166, was designed as a portable and versatile module for operation of radiation detectors. Besides featuring the pulse-height analysis required for spectrometric applications, the device includes detector and pre-amplifier voltage supplies and is designed to sustain battery powered operation throughout a whole workday. The size and weight are kept at a level such that the module can be used single-handedly. The geometrical footprint was

adapted to the size of the HP 200 Palmtop computer. Based on a body of customized software, the MMCA is a widely used instrument in a lot of safeguards applications [1-3]. Although the design paradigm initially foresaw portable field use, the Euratom Safeguards implemented it also for unattended applications. However, as this device was designed more than 15 years ago, a lot of components have become obsolete and increasingly difficult to procure such that the MMCA is approaching the end of its product life cycle.
Therefore, the task was to design a successor to MCA166, which can replace it in all applications, but also take advantage of the increased capabilities of state-of-art components currently available on the market. The development efforts have been undertaken by the company GBS Elektronik GmbH (www.gbs-elektronik.de) located near Dresden/Germany with funding under the German Support Programme to the IAEA. Preservation of the MMCA form factor, power consumption and ease-of-use were formulated as the high priority design features as these are relevant for the MiniMCA as a portable module during in-field verification activities.

The result of the development effort is the digital upgrade to the MiniMCA – the DMCA (or MCA 527) – as displayed in Figure 1. The DMCA integrates pulse-height analysis electronics, high voltage supply for the detector, power supply for the pre-amplifier, chargeable battery packs as well as a control unit and communication ports for connection with a PC.

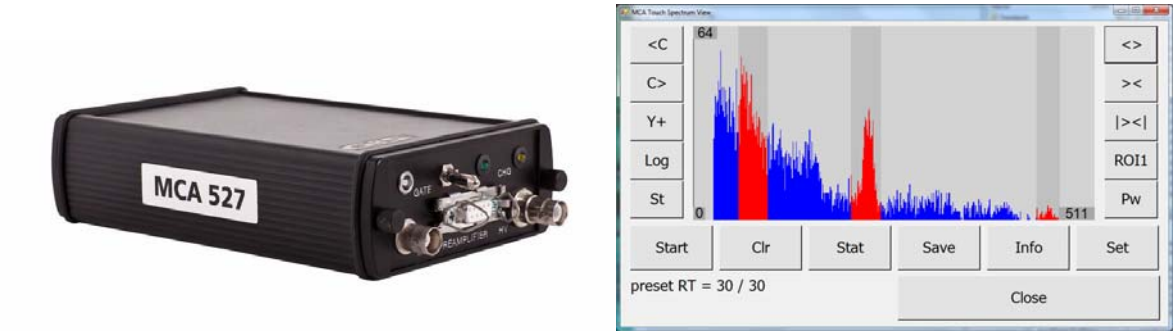

Fig.1: Left: the new digital upgrade of the Mini-MCA (MCA527). Right: Main window of the software MCAtouch designed for interaction with the MCA using a portable touch screen PC.

With regard to the transition to a digital MCA, analogue-to-digital conversion by ADCs with 10MSps sample rate and 14 bit resolution together with a selectable input amplifier and subsequent evaluation by a digital signal processor (DSP) was chosen, as it fully covers the needs for performing a broad range of NDA applications in safeguards verification activities. The DMCA can be used with detectors covering the range from low resolution (NaI, LaBr) via medium resolution (CZT) to high-resolution gamma spectroscopy (HPGe) or in neutron counting applications. The capability of pulse-height analysis is supplemented by the acquisition of spectra in the multi-channel scaling mode and list-mode acquisition of time-stamped data.

Portable computers have undergone enormous progress in the last decade, where mass data storage and touch-screens are now available at relatively low costs. As a tribute to these developments, a software interface for touch screen computers called “MCAtouch” is being designed for control of the MCA and visualization of acquired data. The software is compatible with both MiniMCA and DMCA and includes user-friendly control of measurement tasks (e.g. attribute testing, enrichment determination, criticality test) via the touch-screen. As the field of personal computers is quickly progressing, portability to other portable devices was considered during development of the user interface. As for the analogue MiniMCA, all software commands can be called via a dynamic link library (DLL) which allows users to develop their own application software or integrate the device for example into automatic data acquisition packages [4]

With the intelligence now integrated in the programmable signal-processing unit, upgrading the firmware can open up additional features. Autonomous and automated operation as well as storage of single events in list-mode is a viable option as the MCA-527 is equipped with 2 GB non-volatile memory. Another appealing feature could be the implementation of digital signing algorithms on the DSP for authentication of collected safeguards data.

2. Technical Approach

As the new design is based on digital signal processing, the hardware of the DMCA was newly designed from scratch. The approaches chosen for the DMCA have already been described in a previous publication [5]. Within the scope of this paper, a general overview over technical parameters is given with a focus on a few specific characteristics.

2.1. Signal Processing

The core of the DMCA consists of the ADC which digitizes the analogue detector signal and the signal-processing unit for the pulse shape analysis. The only analogue components preceding the digitizing ADC are a low pass filter, and input range offset DAC for correction of small preamplifier offsets and a coarse amplifier. The coarse amplifier steps are a legacy from the MMCA, but suitable to match the ADC to different preamplifier signal ranges.

Once the matched input signal has been digitized, the pulse shaping for evaluation of the detector input signal is accomplished by digital filtering techniques. Basically two different filters are applied: first, a timing filter which is continuously applied to the digitized pulse in order to sense detector events in the input signal stream. In case a valid event is found, the second filter – shaping filter – is applied for evaluation of the input pulse height. For triggering, single and double differentiating filters are selectable as there is a trade-off between dynamic range and resolving power of pulse pairs. The shape analysis is performed by digital filtering with adjustable rise and flattop times depending on the detector application.

The pile-up of events occurring within a few µs is identified by multiple triggering within the shaping time of the digital shaping filter. As such pile-up events deteriorate the spectroscopic resolution these events are discarded by the so-called pile-up rejecter (PUR).

The technical specifications are summarized in the following list:

Amplifier:
- Coarse amplifier prefilter with amplifications in steps of 1-2-5-10, corresponding to a full scale ADC
- Input signal positive or negative
- Linearity ≤ 0.1%

ADC:
- 14bit, 10 MSps
- Integral Nonlinearity ≤ 0.05%
- temperature stability TK 50

Digital signal processing:
- double differential trigger filter, or single differential low energy low count rate trigger filter
- Pile-up-suppression, pulse pair resolution ~400ns, depending on trigger filter
- Automated and manual adjustment of trigger threshold
- Base Line Restorer with adjustable averaging
- Automated PZC adjustment, detector decay time constants from 33µs to 1ms can be compensated
- Channel splitting 128, 256, 512, 1k, 2k, 4k, 8k or 16k
- Differential nonlinearity <1% for 4k channels and 2µs peaking time

2.2. Technical Specifications

The MCA 527 features the connectors for detectors on one face of the module, and the connectors for charging and communication ports on the opposite face. A computer can be connected via RS-232, USB, or via an Ethernet port for connection to a network. The design of the interfaces (hardware and software) guarantees full compatibility with the Mini-MCA allowing for an easy replacement of the predecessor MCA 166 model. As a compromise to size constraints, only unipolar HV supply units are available, which in case the HV-polarity needs to be changed can be easily replaced without requiring

any tools. The weight amounts to 830 g with a slightly larger footprint than the Mini MCA. Further specifications are as follows:

High voltage:
- Detector HV up to ±3.6kV, polarity change by simple module change

Power supply:
- Li-Ion batteries, operation time 10-25h, depending on detector

Computer Interface:
- USB, RS-232 (38.4, 115.2, 307.2 kBd and 3MBd), Ethernet
- Housing 164 mm x 111 mm x 45 mm without connectors; weight 820g
- Temperature range, operational: at least 0 - 50 °C, limited by batteries
- Humidity up to 90%, non-condensing, IP42

Selection of software features
- System dead time and count rate indication
- Dead time correction
- Automated spectrum recording
- Peak stabilization
- Basic analysis functions (energy calibration, FWHM, peak area and integral calculations, spectrum stripping and smoothing)
- Analysis menu: Energy calibration und further analysis functions according to the purpose. Energy calibration with linear calibration curve using 2 peaks or energy channel pairs

2.3 Additional features

NaI crystals strongly suffer from temperature dependence, and gamma peaks used for evaluation are often subject to temperature peak drifts. Besides logging the internal temperature of the MCA-527 an external temperature sensor signal can be fed into the MCA through an analogue input available at the preamplifier 9-pin connection port for continuous logging. Furthermore gated operation allows separate recording of light pulses provided by crystal detectors with additionally built-in LED stabilization for automated temperature peak-shift correction. Such measures increase the stability of gamma spectrometric applications, which is of particular importance for in-field NDA of uranium .

With the so-called oscilloscope mode the directly digitized input signal can be displayed on the screen of the piloting computer, which provides the possibility of inspecting the detector signal ‘on-line’ for troubleshooting applications.

For environments with strong low frequency (e.g. 50Hz) electronic noise sources, a triple differentiating trigger filter is available for very efficient low-frequency noise rejection. This option is applicable for high-resolution spectroscopy applications up to medium count rates.

The MCA-527 has an internal storage capacity of up to 2 GB non-volatile memory, which allows for implementation of autonomous and automated measurement modes. Additionally, list-mode storage of detected events is now possible for later evaluation of events.

3. Qualitative and quantitative assessment at higher count rates

At higher counting rates, the processing time of the MCA increasingly affects system performance i.e. the number of counts per second which can be processed by the pulse-height analysis is limited by the dead time. Additionally, the high load of pulses leads to deterioration of spectral resolution, as the pile-up leads to erroneous evaluation of the incoming pulses. Therefore, there are two remedial options: (1) reduction of processing time in order to reduce the fraction at which the signal processing is busy, (2) a better pile-up rejection in order to reduce the influence of pile-up in the recorded spectra.

For quantitative determination of gamma radiation peak intensities, an accurate assay of system dead time is crucial for determining the correct live time of the system. A number of approaches are discussed in the literature [6,7]. The approach used for live time correction for steady count rate meas-

urements will be presented in the current chapter along with a measurement to test its performance and limits.

3.1 Pile up rejection and evaluation of the trigger filter time resolution

In analogue MCAs the main contribution to processing dead time is the fixed conversion time of the ADC in the range of several µs. With the digital filtering techniques presented here, the dead time mostly depends on the applied filter length.
Pile up happens when two events happen too close to be evaluated and the energies are partially or fully added. It can be easily seen as background right of a peak and sometimes as pile-up peak at exactly double energy. The approach to solve this problem is to evaluate the signal by two filters: (1) a slow spectroscopic filter optimized for highest precision in determining pulse height and (2) a fast trigger filter optimized for best time resolution. If the fast filter detects events too close to be properly evaluated by the slow filter, they are rejected. The remaining pile up effects at high count rates can be attributed to the still limited time resolution of the trigger filter.
Generally, there is the choice to increase the throughput of the system by using shorter shaping filters. However, shorter shaping filters increase the noise sensitivity which leads to deterioration of spectral resolution. Another option is optimization of the trigger filter, which can be used to improve the pulse-pair resolution of the pile-up rejector. However, there is a trade-off between time resolution and dynamic range, as the fastest trigger filters exhibit increased noise, and designing a more sensitive trigger filter makes it slow. The DMCA offers a selection of trigger filters to optimize a system either for dynamic range or better pile up rejection.

The ability of detecting subsequent events depends on the selected trigger filter and can be evaluated on-line by the analysis of the timing of rejected events as shown in the spectrum displayed on the left side of Figure 2. Here, all the counting statistics of double events identified by the PUR is shown depending on the time difference between the two occurring events. A method to check this is to record a histogram of the time difference between two subsequent events in steps of the ADC clock. It is a reasonable assumption, that this difference should be equally distributed at least for small times. So, it is easy to attribute the missing events at the lower end to the limited time resolution. With standard settings of the trigger filter, pulse-pair resolutions of approximately 400 ns can be reached.

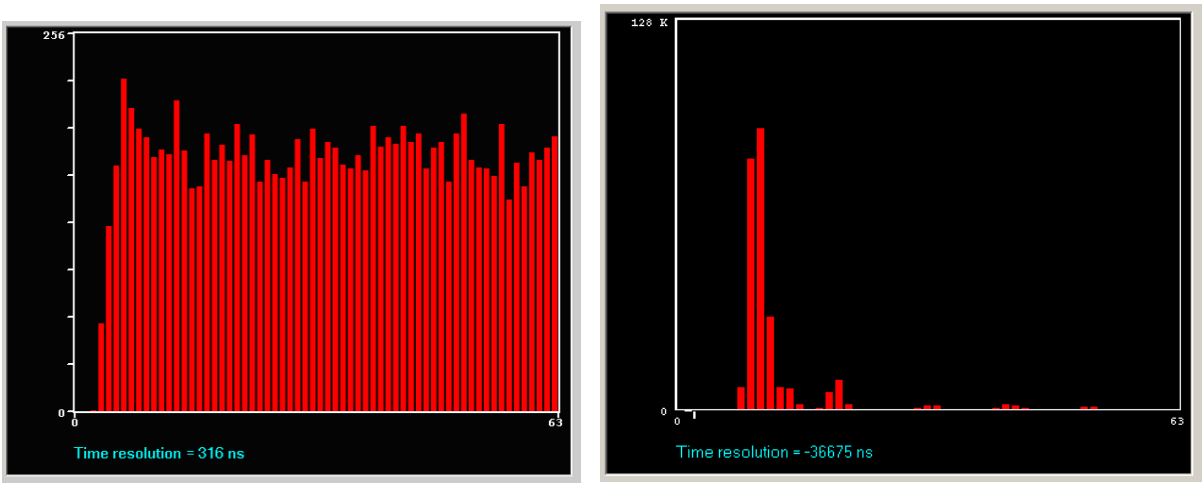

Fig 2: Histogram of the time difference of all events, which were rejected by the pile up rejector, in steps of 100ns and in the range 0..6.3µs. Left: reject histogram of a HPGe with 1-21 trigger filter. Pulse pair resolution is in the order of 300 ns. Right: unsuitable preamplifier signal with ringing and double triggering.

This method can also be used to check if the preamplifier signal is suitable and if the pile-up rejector works correctly. Double triggering because of ringing can be easily seen and otherwise may stay unidentified (see right pane of Fig. 2).

3.2 Dead time correction

Several approaches have been tried to evaluate and correct for dead time; in the end the following approach yielded best results and was implemented:

- Add up times not evaluated $T_{discarded}$, overflow times $T_{overflow}$ and times with trigger filter above threshold $T_{>Triggerlevel}$ and calculate from this a corrected input count rate $\dot{n}_{real}$.
- Calculate dead time from known filter dead time (flattop time t_{flat} +2*shaping time $t_{shaping}$) and input count rate $\dot{n}_{real}$; do this every second.

$$T_{totfast} = T_{discarded} + T_{overflow} + T_{>Triggerlevel} \qquad \dot{n}_{real} = \frac{n}{T_{mess} - T_{totfast}}$$

$$T_{totslow} = T_{discarded} + T_{overflow} + \left(T_{mess} - T_{discarded} - T_{overflow}\right) \times \left(1 - e^{-2\dot{n}_{real}\left(t_{flat} + 2t_{shaping}\right)}\right)$$

T_{mess} is here the real time of the measurement, $T_{totfast}$ is the dead time of the trigger filter only and $T_{totslow}$ is the real dead time of the measurement, also indicated by the software. This approach looks simple compared to other advanced approaches to dead time correction as loss free counting or Zero Dead Time [6], but it works quite well, even with long shaping times and resulting dead times up to 90%.

3.4 Measurements with high input count rates

A standard procedure to test the live time correction accuracy is to perform a two-source experiment. One source (typically the weaker one, but with higher energy, e.g. Cs137) is kept at a fixed position relative to the detector, whereas the other one (typically with lower energy, but stronger, e.g. Co57 or Am241) is located at variable distance to the detector to adjust different count rates. The performance at high input count rates is assessed through evaluation of the peak resolution of the high-energy source and dead time of the MCA (or equivalently the throughput), when increasing the count rate from the low energy source. Ideally, the peak resolution should not be degraded with the PUR turned on and when increasing the count rate. The throughput is of interest as it determines the real-time needed for the measurement. If the live time correction is accurate, the peak area of the first source will always stay the same with same live time and not depend on the total input count rate.

For the DMCA the system throughput is expected to be higher than for the Mini-MCA, as the digital filters are more efficient than the shaping amplifier and ADC conversion of the Mini-MCA. The result of a comparative Am241/Cs137 two-source experiment (HPGe detector) clearly indicates strongly reduced dead-time and great improvement of the throughput of the DMCA (Fig. 3). The live time correction was tested in a further experiment: using a NaI detector, the photopeaks of the two Co60 gamma transition as high energy reference peaks were evaluated at varying count rates with respect to a low gamma energy Am241 source. Spectra at constant live time were recorded for input count rates varying from 2 kcps (Co60 source only) reaching 250 kcps. Within the region-of-interest (ROI) of the selected Co60 photopeak, the variation of the peak integral was below 5 % up to input count rates of 100 kcps and a dead time of 35 %. At higher count rates the approach reaches its limits, and the peak integral of the Co60 reference peak gives unreliable results.

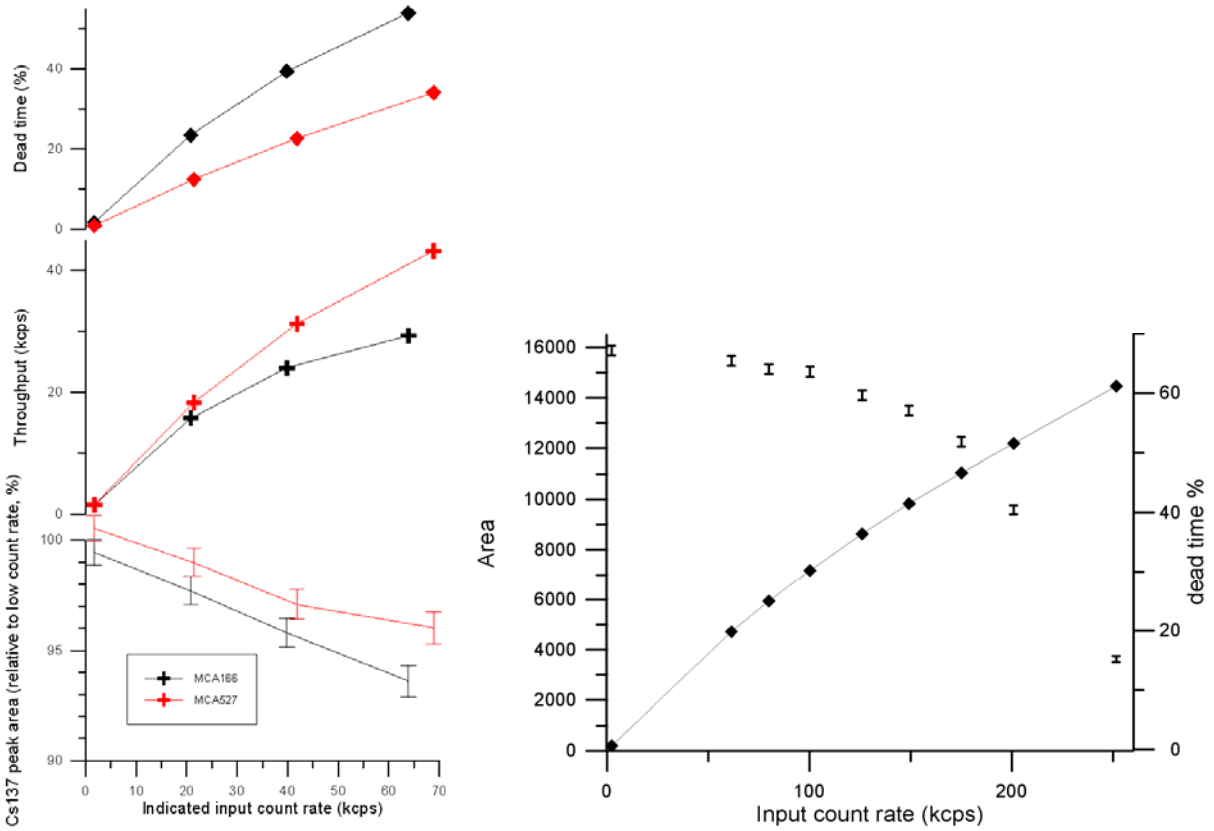

Fig.3: Left: Comparison between MCA527 and MCA166 for a Cs137/Am241 experiment. Throughput, dead time and peak rate error as a function of count rate input. Settings were 1µs shaping time, with MCA527 additionally 1.2µs flattop, long double differential (10-201) trigger filter. Right: Similar experiment, but with NaI detector Amcrys, 0.2µs shaping time, 1.4µs flattop, short double differential trigger filter (1-21), 2kcps Co60 for control, Am241 as load.

4. Measurement of Pu samples with a HPGe detector

Furthermore, it was decided to check the performance of DMCA (MCA527) for Pu isotopic composition measurements evaluated by using the MGA code in comparison whith the results gained with the Mini-MCA (MCA166). The performance criterion was chosen to be the estimated error of MGA for the Pu240 content, as its evaluation is most difficult and sensitive to bad resolution. Similar experiments have already been performed in a previous study [8].
As source a sample of reactor grade plutonium (61% Pu239) was used together with a 1mm thick cadmium absorber. By varying the distance, input count rates from 1kcps to 160 kcps could be achieved. The channel resolution was set to 75 eV/channel.
For both MCAs, 1µs shaping time was used and as measurement time 600s real time was chosen. For the MCA166, the channel setting was 4096 channels and 20*0.8098 amplification . For the MCA527, spectra with 8192 channels were recorded with 10*0.8558 amplification , 0.8µs flattop time of the shaping filter and 4 channel averaging base line restorer (BLR4). Primarily a Canberra GL2010R detector was used, both MCAs were connected to the double output of this detector. As can be seen in Fig. 4, the MCA166 yield a slightly better resolution, however, the MCA527 exceeds its predecessor in throughput by up to a factor of two. A closer evaluation of the peak resolution of the 208 keV photoline results in an evaluated peak width of 700 eV (FWHM) for the MCA166 and 750 eV for the MCA527. Here, settings of the digital filter of the MCA527 can be further tuned as a final optimization step.

However, the relevant performance value is not the resolution of spectral peaks but rather the result gained from using the MGA code for the determination of the Pu 240 isotopic ratio of the measured sample. In a practical application, the measurement error depends on the counting statistics, such that the increased throughput favours the DMCA. Taking in this case the Pu240 error as criterion, the MCA527 shows a better overall performance. Therefore, the twice as large throughput of the MCA-527 compared to the Mini-MCA seems to have more impact than its slightly lower resolution. A com-

parative study of the Pu-240 error as a function of throughput is displayed in Fig. 5: (1) a comparison between two different detector models (Figure 5 left) and (2) between the MCA 166 and MCA 527 (Figure 5 right). The increased performance with smaller errors over a larger range of throughput for the digital upgrade is clearly visible.

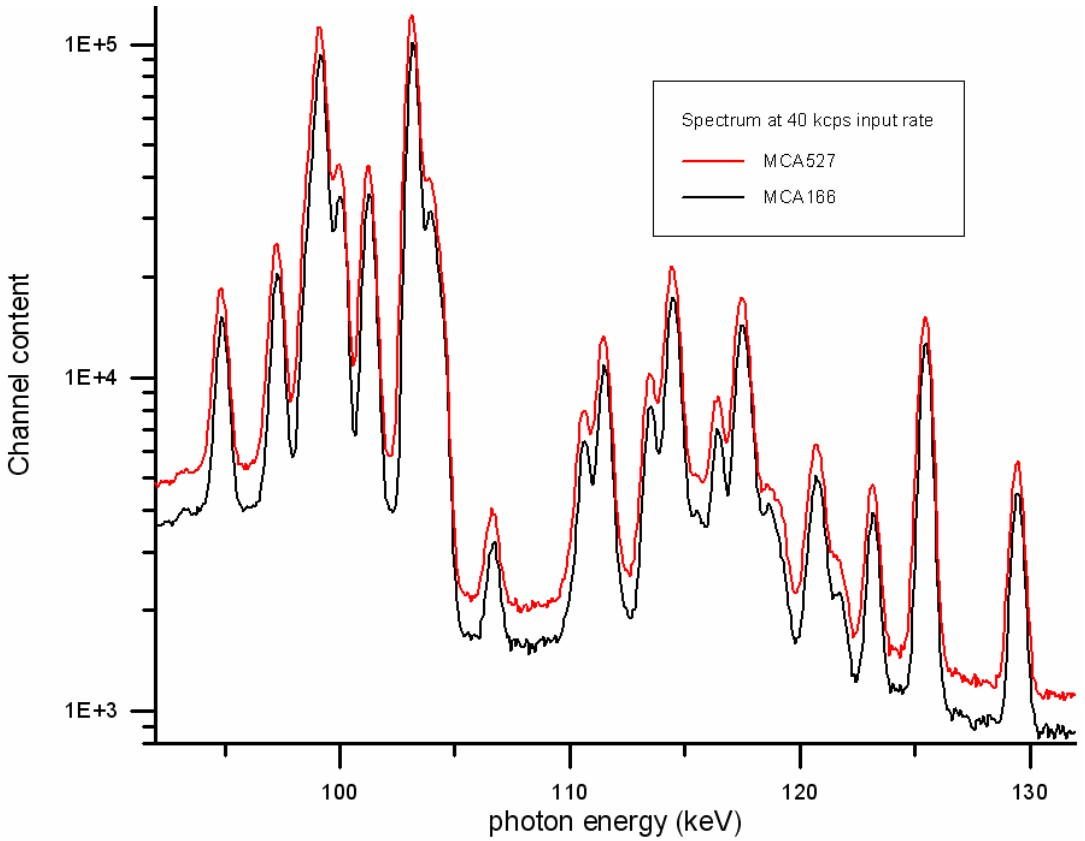

Fig 4: Pu spectrum in the 90-130 keV energy region measured at 40 kcps with 600s real time simultaneously with different MCAs.

Further observations made during the experiments are as follows:

- There was no difference in resolution and hardly any in lower threshold for the MCA527 whether choosing coarse gain 20 and 4k channels or coarse gain 10 and 8k resolution. So, the latter was selected, as with this also the 300 keV energy region of Pu could be used for evaluation.
- With highest count rates, PZC adjustment becomes extremely critical. Settings which were optimized at lower count rates need to be readjusted at higher rates.
- With other HPGe detectors, also lower resolution was observed with higher count rates. As electrical resolution of MCA527 with signal generator signals is significantly better than MCA527, it is worth to have another look at the digital filters and its adjustment.
- Additional higher energy components in the spectrum have an adverse effect on the resolution at lower energies.

The highest input count rate for suitable evaluation seems to be in the order of 100 kcps. Above, there is too much pile up in the spectra and the resolution becomes progressively worse, although up to 250 kcps the input signal will still be processed.

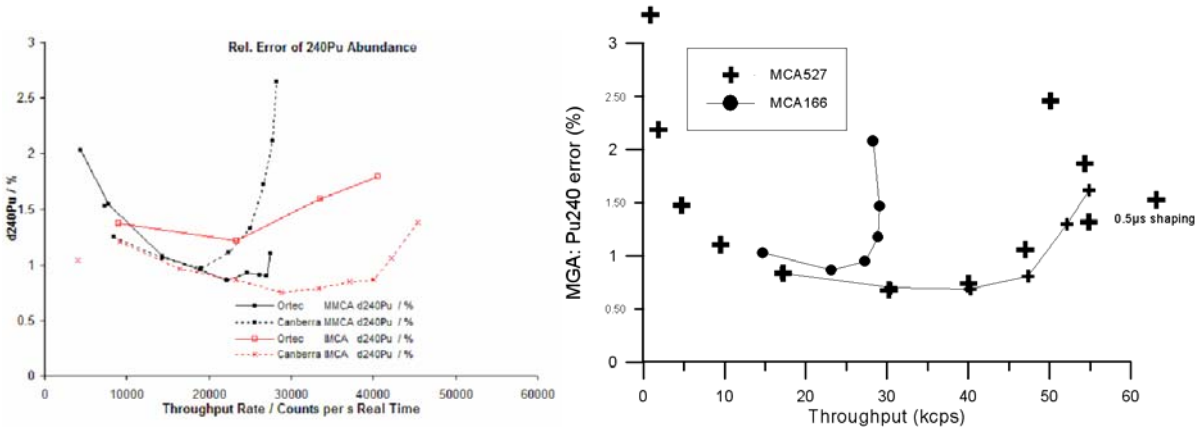

Fig 5: Pu240 error vs. throughput. Left: A comparison of various detector models from Canberra and Ortec. Right: Comparison between MCA 166 and MCA 527. The input count rates for MCA166 were here from 20 kcps to 120 kcps, and from 1kcps to 160 kcps for MCA527.

5. Usability of MCA527 for neutron counting applications

To check the usability of the MCA527 for counting applications, the input of the MCA527 was connected to the output of a neutron counter for coincidence counting applications. This detector provided TTL pulses with a width of 100ns. By virtue of the built-in oscilloscope mode, the digitized input signal can be displayed on the screen of the PC connected to the MCA to analyse the waveform.

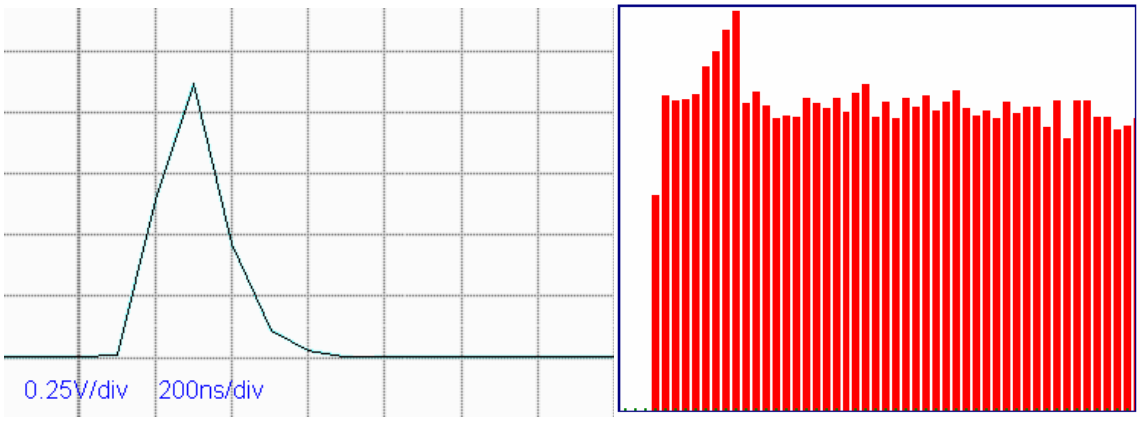

Fig. 6: Left: 100ns TTL pulse from neutron detector, as seen by the MCA527. Right: Histogram of time distances between subsequent neutron events in the range 0-6.3µs, in steps of 100ns.

As the input bandwidth of the MCA is limited to 3MHz, the 100ns pulse gets broadened and the amplitude reduced to around 1V. Nevertheless, this signal is suitable to be counted, as the thresholds can be set arbitrarily. To check the performance, a histogram of the time distance of subsequent pulses was recorded (Fig. 6 right).

This first test shows that the pulse pair resolution is in the order of 0.3µs. More work is planned, e.g. optimisation by threshold selection will be explored.

At the moment, test software exists which allows to record a list of times between subsequent pulses. 32Mcounts are possible due to MCA memory. More sophisticated evaluation software is in preparation.

6. Conclusions

The development of the DMCA is at a stage in which it is ready to be used for measurement tasks. At present, the hardware design has been frozen and the DMCA is already in its production phase. The

digital upgrade of the MiniMCA preserves the functionality of the widely used Mini-MCA. Making use of digital filtering technology increases the throughput by a factor of two, and therefore reduces the real measurement time as compared to its predecessor model. However, some work may still be spent on fine tuning of the digital filters for highest resolution. The feature of peak stabilization for correction of temperature drifts improves gamma spectroscopic measurement results, e.g. when determining the uranium enrichment.

The architecture of the MCA527 has shown its versatility, allowing for much more than the mere measurement of spectra. Further work will concentrate on extending the scope of measurement applications by extensions of the firmware and further development of software.

The developement of a touchscreen user interface is nearing its completion.

7. Acknowledgements

The work is sponsored under task A1791 of the German Support Programme to the IAEA.

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04 EURATOM safeguards

IAEA - EC Cooperation on common implementation of Remote Safeguards Inspections in Europe

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Abstract:

In the current evolution of IAEA safeguards, Remote Safeguards Inspection (RSI) is seen as a tool which will allow, in some specific cases, to increase the efficiency of IAEA safeguards by relying on technical means and on a reinforced cooperation with the Regional Safeguards Systems, State authorities and nuclear operators.

The IAEA and the Nuclear Safeguards Directorate of the European Commission have designed and tested RSI inspection schemes intended to be commonly implemented in the EU civil nuclear installations.

The EU nuclear facilities chosen for field trials are a MOX fabrication plant storage facility, an on-line fuelling power reactor and a Pu store.

The design of the RSI schemes had to be tailored to each of the chosen facilities. Remote monitoring, containment and surveillance, in-line NDA, use of operating data are among the many factors which were taken into account. An additional element of the implementation of a RSI scheme is IAEA unannounced inspection which was also tested during the field trials.
The underlying concepts, the adopted criteria for assessing the outcome of the tests and the very results are presented and discussed.

Keywords: IAEA; EURATOM; Remote Monitoring; Integrated Safeguards

1. Introduction

The Remote Safeguards Inspection project was initiated by the IAEA's DDG Safeguards in late 2009. The purpose of the project was to investigate possible new safeguards approaches that would make use of unattended monitoring, remote data transmission and enhanced cooperation with SSAC/RSAC to improve and optimize safeguards departmental operations and capabilities in order to effectively carry out its verification mission.
The project tested existing technologies and equipment at selected locations, combined with new conceptual safeguards elements, including further participation of SSACs.

1.1. Evolutions of Implementation of safeguards

The Additional Protocol (AP) has been a major factor in the continuing process of strengthening the effectiveness and improving the efficiency of Agency safeguards. It has allowed the IAEA to develop of a conceptual framework for a system that “integrates” the new measures under an AP with the established measures under a comprehensive safeguards agreement. Integrated safeguards are based on a set of concepts, approaches, guidelines and criteria that govern the design, implementation and evaluation of this system. This framework has been designed to ensure consistency in implementation, and to provide the flexibility to take account of State specific features and characteristics in order to optimize effectiveness and efficiency.

Under this system routine safeguards implementation does not discriminate between States. All credible paths to the acquisition of weapons-usable material are covered by safeguards measures but a conclusion of non-diversion is never-the-less based on nuclear material accountancy. One major new element of this safeguards system is a conclusion of the absence of undeclared nuclear material and activities which is drawn and supported by comprehensive information review and evaluation for each State.

1.2. Limited Resources

Strengthened safeguards have an increased requirement for information collection and analysis, coupled with the continued traditional safeguards inspection activities, within a constrained resource and budget environment of the Agency. Integrated safeguards utilises the extension of timeliness goals for certain nuclear material and the use of random inspections to achieve inspection efficiencies whilst still retaining effectiveness, but it has been recognised that more savings could be achieved.

1.3. Repetitive Verification Tasks

Inspection activities consist of the verification and evaluation of accountancy data, nuclear material measurements and containment and surveillance results. In many facilities there are few, if any, changes in inventory or status for several months and the inspection activities are limited to data collection from installed safeguards seals or other equipment. These repetitive activities have normally relied on inspector presence which is both time consuming and expensive.

1.4. Travelling to remote facilities

Travelling from IAEA or Euratom headquarters to nuclear facilities is a substantial part of the human and financial resources invested in the implementation of safeguards regime.
Activities of nuclear facilities include shipment and reception of nuclear materials, transfers of spent fuel inside the facility. Planning of some of these events, namely their starting time and duration, can be only approximate. This can cause a severe strain in the available resources
Also periodical verification of containment and surveillance media (e.g. seals, video recording) has been demanding dedicated and resource-consuming missions for in-situ work performed by IAEA or Euratom inspectors.
For this, by using remote controlled containment, video surveillance and monitoring by neutron- or gamma-measurements, interesting savings can be achieved.

2. Principles of Remote Safeguards Inspection

2.1. Information Driven Safeguards

The move by the IAEA to a system that foresees further emphasis on the analysis of inspection, complementary access and open source information, has been termed 'safeguards that is fully information driven'. Remote safeguards inspections (RSI) could reduce but not eliminate inspectors’ routine presence in the field allowing more time for important inspection and headquarters work. An RSI approach would be an integral part of a state level approach that considers the State as a whole and provides the opportunity to take State-specific factors into consideration in all stages of safeguards implementation

2.2. Enhanced Cooperation with Regional Safeguards Systems

Technology coupled with techniques such as regular advance declarations, random inspections and authenticated operator/SSAC/RSAC activities together form the basis of RSI. The implementation of these activities requires a competent, cooperative partnership between Agency, SSAC, RSAC and facility operator.

2.3. Remote Data Transmission

RSIs could be seen as an extension of existing technologies currently employed, including remote monitoring, unattended mode measurement equipment and remotely monitored electronic seals.The vision that inspectors in the future are able to make better use of their time in the field requires the enhancement of technology that will provide them with extensive and improved data gathering equipment with the capability to transmit data securely and reliably to headquarters where enhanced analysis tools would allow an effective evaluation of facility status. This has to be implemented while complying with:

- a secured handling of the inspection data which will rely on the encryption technology and, when requested, on a delayed transmission which allows buffering of the information for being security-checked before being transmitted to Luxembourg or Vienna
- the avoidance of a extra burden for the operator in term of interference with his operational activities

3. Basis for implementing RSI in EU

3.1. IAEA – EC Partnership Approach

The IAEA and the European Commission are currently implementing nuclear safeguards in the EU on the basis of a Partnership Approach which has been recently developed on the following cooperation principles:

- Principle of ‘observation’ to be replaced by the one-job-one-person supplemented by quality control for all inspection activities;
- Use of common approaches and procedures;
- Commonly shared analysis of DA samples;
- Use of common instruments and techniques;
- Cooperation in research and development and common training of inspectors;
- Use of the same basic parameters for defining the extent and frequency, such as significant quantities, detection probabilities and timeliness goals.

Different types of internal documents were agreed between EC and IAEA for the practical implementation of common inspection and support activities.

3.2. Common Use of Equipment

In the framework of the Partnership Approach and under the supervision of the EC-IAEA Liaison Committee, the IAEA and the EC have drafted common technical procedures which provide guidelines on the following subjects:

- Installation of joint-use IAEA/EC surveillance systems;
- Surveillance Review Procedure;
- Joint Use Arrangements for IAEA/EC EOSS Seals;
- Spent fuel cask loading/sealing procedure;
- Sealing arrangements for joint-use surveillance equipment;
- Arrangements for Joint-Use Remote Data Transmission.

In the case of the design of RSI trials the above mentioned procedures have been used as a consolidated reference for both organisations.

3.3. Common Training

Common IAEA and Euratom inspectors training has been recognised by the DDsG of both organisations as a priority for the implementation of international safeguards in the EU. New courses on communication and team work have been run recently with full satisfaction of all the delegates. Common training is a factor that helps considerably in having IAEA and Euratom inspectors carrying out joint inspection activities smoothly and efficiently.

4. Field Trials in EU facilities

The selection of facilities was based on several factors, derived from the objective to reduce the inspector effort in the field in order that more effort is available at headquarters for analysis work. Therefore an expected reduction of inspection effort at the facility was one of the determining factors.

The field trials were expected to take place at facilities which already had a combination of surveillance, electronic seals or nuclear material measurement equipment already installed and in use. Operational remote monitoring for some of the equipment would be an advantage.

The final, and probably most important, requirement was that of a cooperative and supportive SSAC/RSAC with a high knowledge of safeguards, good quality of activities and strong communication with the IAEA.

Potential field test facilities were discussed at length with SSAC and RSACs leading to the final selection of a plutonium store in the United Kingdom, a MOX fabrication plant in France and an on-load reactor in Romania which were to be tackled as a joint project between the IAEA and the EURATOM.

The three trials would be able to test the combination of mailbox system declaration, unannounced visits, random inspection and operator/SSAC activities on material measurement and electronic sealing.

4.1. Sellafield Pu store QS19

The Pu store in Sellafield QS19 is a Material Balance Area (MBA) where large quantities of nuclear material are stored. The frequency and number movements in the store are foreseen to be limited; this situation is favourable to the adoption of sealing as method for preserving the continuity of knowledge.
Sealing can be implemented by using long loops of optical fibres which are connected to electronic seals. The used seals have the capability of sending data remotely.
Video surveillance complements the C/S scheme of the MBA. Both seal and video data are remotely transmitted to the Euratom and IAEA headquarters.
Procedures, based on operator mailbox declarations of access requirements, have been developed to allow operator-alone access to un-seal and re-seal the main store access doors without inspector presence.

4.2. Melox MOX FFP

The type of activities targeted by the RSI trial are related to the shipment of fresh MOX fuel shipped from Melox to non-French reactors. The operations of NDA measurements, transfer to the transport containers and sealing have been foreseen as sending the related data to local loggers which can be reviewed during a random or a planned inspection.

4.3. Cernavoda NPP

The Romanian CANDU reactors in Cernavoda are safeguarded with the help of neuron and gamma monitors which cover internal flows of nuclear material.
Electronic seals are also used for keeping the continuity of knowledge in the dry store areas. The devices indicated above have been used for the RSI trial and have been sending data remotely to the IAEA and Euratom headquarters.

5. Common IAEA – EC evaluation criteria

5.1. Definition of RSI field trials

For each chosen facility, the definition of the overall purpose of the trial focused on potential for improving the overall safeguards approach of the facility, especially in terms of savings in manpower for each organization.

The following points have been identified as basis for the design of an RSI scheme:

- Targeted materials and locations in the facilities, items and their identifiers, movements to be detected and any other action designed to ensure "continuity of knowledge" (CoK);
- Equipment (including software) to be used, either already in place or to be added;
- Identification of the type of information and quantification of the amount of data to be remotely transmitted;
- Define in detail the measures needed for validation purposes (data authentication and encryption, use of sealed calibration sources, coupling of electronic seals and video surveillance, etc);
- Definition of the distribution of tasks between operator and inspectors form both organisations;
- Quantification of the expected effort for the trial.

Any trial had to be based on a coherent set of information provided by the plant operator. For this, IAEA and Euratom for each trial had to:

- identify the operator's data (formal declarations, operating data) to be used for the RSI scheme, investigate whether additional operator information would enhance the quality of the RSI scheme and;
- define the way the information is handled while complying with security rules.

5.2. Planning of RSI field trials

The concerned national authorities and operators have been contacted by Euratom and the IAEA to explain the details of the trial and to agree on the suitable week for carrying out the trial.

A global assessment of the demand of both human and technical resources was determined for all the test cases foreseen in the EU.

Trial activities have been carefully planned in order to fit into both IAEA and Euratom already established safeguards activities, without jeopardizing current safeguards implementation.

5.3. Criteria for common assessment of the results of the RSI field trials

When evaluating the trial results, evidence had to be brought on the full understanding from both operator and inspectors of the carried out activities and of their respective roles within facility-specific RSI scheme.

As the containment and surveillance measures are an essential part of a RSI scheme, the CoK has to be convincingly ensured through all the trials as well as the proper functioning of the used equipment.
It was important that at the and of a trial the inspectors had time enough to complete successfully the assigned tasks

Other factors were taken into account such as:

- Actions carried out for RSI did not interfere with the planned activities of the operator and of inspectors;
- Communication and coordination with the operator went out smoothly;
- RDT lines were not overloaded and in general the functioning of the installed safeguards technical equipment (both Euratom and IAEA) was not compromised by RSI activities.

6. Results: conclusions and Considerations

The overall result of the field tests has confirmed the validity of the concept of Remote Safeguards Inspections, however it should be recognised that most of the individual elements, including surveillance and seals, non-destructive assay (NDA) measurement and associated data transmission combined with operator/State assistance is not new. The field trials presented a particular combination of the remote monitoring and data transmission schemes developed and implemented specifically at these facilities for optimising the inspection resources

The result of the field tests show that a reduction of inspection effort is possible at the selected field test sites and does indicate that there still exists room for further economies. However, the field tests did not include a full cost benefit analysis, as they utilised existing installed equipment which was subject to already planned routine maintenance. Should the concept be promulgated to further facilities the equipment, installation, infrastructure and maintenance costs should be considered.

The lessons learned from the field trials indicate that further optimisation of IAEA effort is possible by the transfer of the routine in-field activities to headquarters (HQ), early evaluation of information for prompt response and improved effectiveness, and enhanced use of SSAC/RSACs which leads to improved efficiency. It was further indicated that the agreement on remote data transmission and infrastructure must be in place and the in-field activities need extra training for RSAC/SSAC staff, the operator and the IAEA in order for the PDI savings. The limitations of the field trials are: all tests were performed only with the EURATOM, no cost benefit analysis was performed, and the potential misunderstanding of the term “Remote Safeguards Inspections”.

7. The way forward

There are several areas which if tackled could be of benefit to the concept of remote safeguards inspection. The cornerstone of RSI is remote monitoring and therefore an expansion of the number current remote monitoring systems and improvements to the remote monitoring infrastructure is a natural way forward. The upgrading of transmission technologies to enable simultaneous data and surveillance review would bring capacity and opportunities to RSI projects.

Improved authentication of unattended measurement equipment and implementation of advanced systems to allow third party operation and maintenance would further reduce the requirement for routine inspector presence at facilities, as would the development of better methods to authenticate SSAC/RSAC activities, who would then be free to perform additional verification activities, the results of which would be acceptable to the IAEA.
There needs to be continuous improvement of all aspects of remote monitoring and data transmission. The experience gained and the lessons learned from the field trials have been very valuable in the evolution of the State-level concept and a safeguards system that is fully information driven.

Development and use of Remote Data Transmission in Reprocessing plants in the EU: towards Remote Safeguards Inspections?

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Abstract:

Reprocessing plants share with other bulk handling facilities some characteristics which makes them ideal for the installation of unattended systems for safeguards purposes.

In the EU reprocessing plants such unattended systems, either purpose designed and built Euratom proprietary equipment, or the branching of operator's devices signals, were installed already in the late 1980's and further developed in the subsequent decades. Signals from unattended systems are normally transmitted to the inspector's office on site for further treatment, analysis and comparison with the operator's declarations.

In recent years the efficiency of the EURATOM safeguards system has been drastically increased by re-locating a substantial number of the controls and checks from the sites to HQ in Luxembourg. This has become possible by obtaining the agreement of operators and State Authorities for the transmission of nuclear safeguards measurement or monitoring data to Luxembourg.

Remote Transmission of all Data (RDT) relevant to a safeguards inspection is seen by Euratom as the only way to improve the security of the transmitted data and facilitate their archiving and effective exploration at HQ. Meeting RDT requirements could also indirectly contribute to Operator's further stratification and consolidation of data provided.

This paper also addresses the issue of the relation between RDT, Remote Safeguards Inspection (RSI) and possible reduction of Inspection Mission Days (IMDs). The position taken is that RDT could be used so that inspectors (and inspection equipment) are better prepared at the onset of an inspection, which will allow them to perform less repetitive tasks while on site while concentrating on BTC or supporting documents verifications or audit of operator's systems.

On the other hand, RDT could be an important element of RSIs especially for large static stores for direct use nuclear material equipped with effective containment and surveillance: it allows inspector and operator dose uptake reduction and increases the flexibility of inspector and operator maintenance interventions in safeguarded areas.

Keywords: Remote Safeguards Inspections, Remote Data Transmission, Nuclear Safeguards

1. Introduction

Reprocessing as a back end option was already introduced during the early conception of the nuclear fuel cycle of many EU Member States. As a result, a number of Member States embarked on the development of reprocessing technologies, the construction of demonstration and pilot plants, and eventually in the setting up of commercial operations . The plants in Sellafield (Magnox and Thorp) and in La Hague (UP2 and UP3) are in commercial operations since a number of decades, and represent more than half the world wide installed reprocessing capacity.

Reprocessing plants are characterised not only by their complexity and difficult access in many areas due to high radiation levels, but also by their continuous mode of operation requiring a high degree of automation and the presence of significant amounts of direct use material both as throughput and inventory.

As a consequence of the above characteristics of reprocessing plants, safeguards approaches based on the installation of unattended equipment operating in continuous mode, maximum use of containment and surveillance systems and most important, transfer of the signals/information collected either to a dedicated technical room or preferably to the Inspectors' office on site were conceived already in the 70s and 80s [1,2].

The existence of such central data collection configurations was further exploited at a more recent stage, with the remote transmission of some of the data collected towards the Euratom Headquarters in Luxembourg, and was complemented by the electronic transfer of some of the operators operating data to Luxembourg, making thus the comparison between operating records, inspectors' measurement observations and results, feasible at HQ .

This paper will attempt to describe the current state as far as remote data transmission of safeguards related data from Reprocessing plants to the Euratom HQ is concerned, their possible use at headquarters in order to reach safeguards conclusions, and finally the role of RTD in increasing the efficiency and effectiveness of the safeguards system via the introduction of Remote Safeguards Inspections.

2. Current Safeguards Approaches in Reprocessing Plants

Safeguards approaches in Reprocessing Plants are developed after careful examination of their basic technical characteristics, taking into account that after the active start up of the installation some activities (e.g. the accountancy tank calibration or the verification of absence of undeclared circuits) could not be realised without undue irradiation burden on both the operator and the inspectorate. An early start of the discussions between the operator and Euratom and the inclusion of some Euratom safeguards requirements in the design of the plants, is essential.

The first step in designing the safeguards approach is the division of the plant in Material Balance Areas and the Introduction of Key Measurement Points, in such a way that the control of the flow of material and its inventory becomes feasible by the operator's accountancy system and can be controlled by the inspectorate.

A typical structure for a reprocessing plant includes the following MBAs:

- Wet storage of irradiated fuel elements
- Shearing and dissolution of elements
- Separation and conditioning of Pu , U and fission products
- Pu product storage
- U product storage
- Waste treatment facility

Within each MBA the safeguards approach comprises the following elements:

- Verification of the flux of nuclear material
- Annual Physical Inventory Verification
- Evaluation of the MUF (in the bulk handling MBAs)

- Evaluation of the C/S measures for Continuity of Knowledge purposes

Depending on the MBA type and the state of the Nuclear Material the following verification activities are performed in order to implement the safeguards approaches:

- Item Identification
- Weighing
- Volume and density measurements of tanks
- Continuous monitoring of the liquid level in the tanks
- DA sample taking and analysis (concentration, assay, isotopic composition)
- NDA measurements (neutron and gamma)
- Neutron monitoring
- Video surveillance and electronic sealing
- Use of Copper Brass metal seals

With the exception of the item identification and the DA sampling, all other verification activities are based on continuously monitoring systems connected to a local acquisition system. Raw data are transferred to the inspectors' office on site, are analysed and compiled to the 'events database'. The latter is then compared with the operating records as declared by the operator.

As it will be explained in the next point, on some occasions the 'events database' is transferred electronically to the Euratom Headquarters in Luxembourg, where it is compared with the operator's operating records and reports, also transferred to Luxembourg by electronic means (USB stick).

Current inspector presence in Reprocessing Plants (number of inspectors and frequency of inspections) is rather related to the bulk of the information provided, the time needed to process it and the requirements of the operator to access part of the installation for maintenance of equipment than any nuclear material timeliness criteria.

3. Development of Remote Data Transmission

As discussed above, a big amount of raw data are recorded by the various measurements and monitoring systems installed at Reprocessing Plants, and are analysed and compiled into the events database to be compared with the operator's operating records and reports. At the same time, a large amount of state of health information is available. The transfer of all above data via commercially available transmission lines from the nuclear installation to the Regulator's Headquarters, is understood under the term Remote Data transmission (RDT).

Data candidate for RDT could be clustered into the following categories :

- Euratom equipment state of health
- Euratom equipment measurement data
- Branching of Operator's equipment
- Euratom video surveillance data
- Euratom electronic seals data
- Operator's operating records
- Operator's operating reports
- Operator's supporting documents
- Inspector's inspection working documents

All above data are considered as sensitive and carry a confidentiality classification at a level that might vary from one Member State to another. It is thus important that for each category of data, Euratom, the State Authorities and the Operator involved, come to an agreement on exactly which data and in which way it will be transmitted so that the security requirements are met. Particular precautions have to be taken when data already classified are put or transferred together (e.g. data pertaining to nuclear material weights, location and time).

So far, for Reprocessing Plants in the EU, Euratom has obtained the agreement of the competent authorities for the remote transmission of some C/S and Euratom equipment data from Sellafield [3],

while Operator's records and supporting documents are transferred using USB sticks from both Sellafield and La Hague.

Details of the technical and security arrangements agreed have been presented on earlier occasions [4] . In order to avoid issues related to the volume of the data transmitted and the synchronisation of the data bases in Luxembourg and on site, the preferred option would be to store all data on dedicated servers on site and access them via remote secure protocols from Luxembourg. The same servers would serve as the depository of the operator's data currently scanned and transferred via USB to Luxembourg.

The advantages of RDT would then be used to their full extend . Such advantages are:

- Increased data security through appropriate secure transmission
- Better possibilities of organising, exploring and archiving the data
- Data consolidation and stratification through early agreement on which data need to be provided
- Better possibilities to develop programs for data analysis and evaluation, based on the agreed format and context of data transmitted
- Increased efficiency through harmonisation and streamlining between facilities of the same kind
- Improved conservation and transfer of inspection know-how and the cross-fertilisation between teams of inspectors

4. Towards Remote Safeguards Inspections

With such a plethora of data remotely transmitted and available at Euratom headquarters, the question of their use towards improving the efficiency and the effectiveness of its safeguards verification activities becomes evident.

Activities that could be performed at Headquarters to support or to prepare inspection activities include the following:

As far as equipment is concerned:

- Follow up of the state of health of equipment and planning of interventions
- Planning of alternative safeguards measures in case of equipment failure or loss of containment
- Remote update of system parameters (mainly software)
- Checking of completion of maintenance or upgrade actions requested by the inspectors (e.g. transducers calibration, or testing of proximity switches)

As a result of the above, the operator would have higher flexibility in planning and performing maintenance operations, and could have enhanced access for maintenance of his/her equipment installed in controlled areas (currently limited during inspector presence). Reduction of dose uptake for the inspectors but also for the operator via better planning, is obvious.

As far as information is concerned:

- Review of surveillance records and reconciliation with events as declared by the operator
- Review of electronic seal events and reconciliation with events as declared by the operator
- Evaluation of continuous measurements (e.g. accountancy tank volume and density, or neutron and gamma measurements) and comparison with operating records.
- Evaluation of unattended NDA measurements of discrete items against the operator's declarations
- Comparison of operating records and reports with accountancy declarations received in Luxembourg

As a result of the above, possible discrepancies will be detected and analysed before the arrival of the inspector to the installation, giving adequate response time to the operator.

The question now arises whether the activities at Headquarters described above could replace the physical presence of inspectors at facilities, and could thus be considered as a safeguards inspection executed remotely.

The answer to this question could be different, depending on the type and characteristics of an installation:

For example, in static stores with effective C/S (e.g. a multilayer containment and ideally a robust, reliable and failsafe outer containment layer (door switch), a number of physical inspections could be substituted by Remote Inspections without either jeopardising the possibility of the operator to access the store for equipment maintenance, or increasing the probability of follow up measures due to C/S failures.

On the other hand, in bulk handling facilities, the physical presence of inspectors would have to be continued at a fairly high level, but the emphasis on their work on site would be shifted to activities like BTC re-verification, examination of supporting documents, audits of the operator's NMAC system, and resolution of anomalies. It should also be noted that physical presence of inspectors familiarises them with the installation and the processes, and is of utmost importance when introducing new inspectors to a new facility. Direct contact with the personnel of the installation should also not be underestimated as a positive factor for problem solving. Inspector presence serves also as a deterrent for potential diversion and promotes application of good operational practice.

Another factor to be considered is related to the optimisation of the inspector's time, both at Headquarters and during inspections. It can be argued that a Remote Inspection Scheme could lead to a more efficient use and a homogenisation of the inspector's work pattern at Headquarters, and an increase of their efficiency during inspections (through better planning and reduction of unforeseen difficulties).

For the operators, a Remote Inspection Scheme could only lead to advantages, related to the flexibility in planning their equipment maintenance interventions and the predictability of the inspectorates' requirements during inspections. Possible reduction of physical presence of inspectors would also reduce the need for the provision of escorts and operational staff for manipulating nuclear material, and would thus have an indirect positive effect.

An issue that will need to be addressed, is the integration of the verification and comparison activities performed at Headquarters to the overall safeguards conclusions reached as a result of an inspection. A procedure replacing the current "opening meeting" i.e. the clarification of the scope of the verifications, the transmission of operating data and the discussion of any current issues potentially influencing the verifications, should be introduced. The format can however be more mechanistic and consist of a functional mailbox and standardised checklists per type of verification. The same applies for the 'final inspection meeting' during which the preliminary results of an inspection are presented to the operator. The latter will have to be substituted by a different means of communication (e.g. videoconference) which will allow the Inspectorate to inform the operator of the preliminary results of the integrated verification activities at Headquarters and on site.

5. Conclusions

Reprocessing plants and Nuclear material stores are well equipped and well suited for Remote data transmission of safeguards related data.

Remote data transmission offers a number of advantages related not only to the better organisation and the security of data transmitted, but also to a better and timely preparation of inspections – reducing thus unforeseen problems.

Introduction of RDT as an element of a remote inspection scheme could be advantageous to all involved. Under such a scheme, the range of inspector's activities at Headquarters and during inspections will have to be revisited so that an optimisation of the efficiency and the effectiveness of the system are achieved without any undue extra pressure on the operator.

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The Role of NMAC Audits in Euratom Safeguards – Development of an audit framework

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Abstract:

The use of audits of nuclear facility operators' nuclear material accountancy and control (NMAC) systems has evolved since the idea was launched some years ago. The European Commission has developed a framework that enables the use of NMAC system audits as an effective and efficient tool in nuclear safeguards. The framework includes elements like audit definition and concept, a procedure, audit criteria and the approach for using audits. The main elements of this framework have been built upon ESARDA working group recommendations and were widely consulted with Member States and nuclear operators. The framework and experience from its application are presented.

Keywords: Safeguards concepts (policies, perspectives, limitations, Strengthened and Integrated Safeguards, State and Regional Systems, Quality Assurance Approach)

1. Introduction

The audit of nuclear operators' NMAC (Nuclear Material Accountancy and Control) systems is a powerful tool in nuclear safeguards. When nuclear operators design and implement a high quality NMAC system, nuclear safeguards processes run in a very efficient way. This allows the nuclear operators to benefit from a minor burden of inspections and facilitates the work of the safeguards inspectorate.

The European Commission has developed and implemented the necessary concepts and procedures to use NMAC audit in order to detect and highlight improvement opportunities in nuclear operators' NMAC systems when necessary. The audit framework follows the model of those created for Quality Management Systems and Environmental Management Systems. These frameworks have been widely accepted worldwide.

NMAC audit is a systematic and independent comparison between a real NMAC system and a high quality model. To make this comparison, latest international standards regarding audit and bodies providing audit services are applied. The European Commission has succeeded in adapting these standards to the particularities of a regional nuclear safeguards organisation.

2. Evolution of the NMAC audit implementation

The implementation of audits of the NMAC systems of nuclear operators was launched in 2004 by Commissioner De Palacio [1]. Further, the Commission Staff Working Document [SEC(2007)293] « Implementing Euratom Treaty Safeguards » (IETS) [2] stated explicitly as a major element in its implementation the development of a baseline reference to be used as a basis for assessment and as model for nuclear operators.

After some previous experiences, in 2006 the European Commission initiated a first series of trial audits in installations of the United Kingdom and France. These trial audits focussed on specific safeguards processes of the nuclear operators. It was necessary at the time to develop tailored audit criteria and

procedures. The conclusion of these trial audits was that the European Commission was capable of performing NMAC audits in an independent and professional way.

The IETS document expressed for the first time in a Commission official document the benefits of getting support from ESARDA. Under the initiative of the European Commission ESARDA set up a Working Group focussed on the development of audits of NMAC systems. The main outcome of this Working Group is a detailed model of a high quality NMAC system and guidelines to conduct audits based on standard ISO 19011:2002 Guidelines for quality and/or environmental systems auditing [3].

Based on the outcome of the ESARDA NMAC Audit Focus WG, the European Commission developed an audit procedure and a baseline reference to be used as audit criteria. The audit procedure and audit criteria are key elements to conduct audits. A second series of trial audits was designed in 2008 to test the utility and completeness of the audit criteria and the audit procedure. This series consisted of three full scope audits in a nuclear power plant, a fabrication plant and a research centre in Spain and Germany. The results of the trial audits demonstrated that the audit criteria covered completely the audited operators' NMAC systems. The audit criteria are usable and adaptable to different facility types. The audit procedure made the complete audit process run correctly.

After a wide consultation with nuclear operators and Member States authorities, the audit criteria were laid down in the form of the Commission Recommendation of 11 February 2009 on the implementation of a Nuclear Material Accountancy and Control system by operators of Nuclear Installations (Euratom/120/2009) [4].

Under the initiative of the European Commission, the ESARDA Audit Working Group started working in 2009 with the main task of making an interpretation of the audit criteria. The interpretation was aimed to help nuclear operators to find solutions to the requirements of the Commission Recommendation (Euratom/120/2009) [4] and help auditors to collect the necessary information during audit activities. The work was finalised by the end of 2010 and it is now ready to be used.

The European Commission has developed the audit concept based on the International Organization for Standardization (ISO) definition of audit. An audit approach has also been defined to identify when the use of NMAC audits make nuclear safeguards more effective and efficient.

3. The audit framework

The European Commission NMAC Audit framework is based on the model of quality management systems audit and environmental systems audits of ISO. The main elements of this framework are the audit concept, the audit criteria, the audit procedure and the audit approach.

3.1 The audit concept

NMAC Audit is a documented and systematic process to compare a high quality NMAC model (**audit criteria**) with the actual implemented NMAC system of the nuclear operators. To perform this comparison, information (**audit evidence**) is collected and differences between the model and the reality (**audit findings**) are highlighted together with improvement opportunities. The process can involve the whole operator's NMAC system (full scope audit) or some of its components.

This concept is fully supported by the ISO definitions of audit, audit criteria, audit evidence and audit findings.

3.2 The audit criteria

The definition of audit criteria according to ISO is a set of policies, procedures or requirements, which are used as a reference against which audit evidence is compared. As mentioned above when performing NMAC Audits, the European Commission will take as audit criteria the requirements laid down in the Commission Recommendation (2009/120/Euratom) [4]. It should be noted that these requirements could be complemented when needed by normative documents (standards, guidelines, recommendations) accepted as latest international standards for a specific item included in the scope of the audit. As an example, the International Organization of Legal Metrology (OIML) Recommendation R-76-1 Non-automatic weighing instruments. Part 1: Metrological and technical requirements – Tests [5] will be considered as audit criteria while auditing the way a nuclear operator uses and controls the weighing machines used for nuclear material accountancy purposes.

The audit criteria describe the main elements that a high quality NMAC system should have and the controls to apply on these elements. The management of the NMAC system, the measurement and measurement control programmes, the nuclear material tracking system, the data processing system, and the material balance activities are described in these criteria.

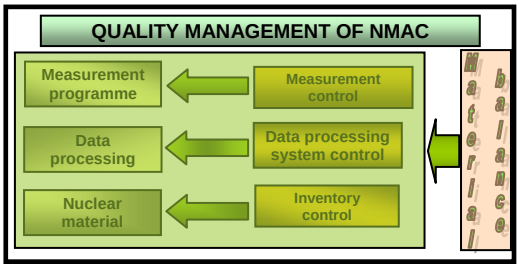

Figure 1: NMAC system model

The way an NMAC system should be managed is based on the most widely recognised quality management systems. It is described in Section 3 of the Commission Recommendation (2009/120/Euratom) [4]. The management criteria stress the need for visibility and relevance of nuclear safeguards in the organization of the nuclear operator. For instance, training of staff involved in the management of nuclear material is recognized as a crucial element of the NMAC system, together with the process approach to be applied to the NMAC activities.

The way measurements should be performed and controlled is described in Section 4 of the Commission Recommendation (2009/120/Euratom) [4]. The criteria reflect in a faithful manner the latest international standards regarding measurement systems as ISO/IEC 17025:2005, general requirements for the competence of testing and calibration laboratories [6], and ISO 10012:2003, Measurement management systems, Requirements for measurement processes and measuring equipment. [7]

Section 5 of the Commission Recommendation (2009/120/Euratom) [4] states the need for a high quality NMAC system to be able to determine at any time the quantities and characteristics of all nuclear materials present in each location of a nuclear facility.

The data processing system to transform operational and measurement data into accountancy data and declarations is described in Section 6 of the Commission Recommendation (2009/120/Euratom) [4].

The material balance activities described in Section 7 of the Commission Recommendation (2009/120/Euratom) [4] are an overall control exercise for the sum of NMAC activities. In this section it is described how receipts, shipments and other inventory changes of nuclear materials should be managed and which quality control and quality assurance measures are needed for effective physical inventory taking and material balance evaluation.

When needed, interpretation of the audit criteria will be made according to the document Applicability and Interpretation of the NMAC Audit Criteria [8]. This document is a support document helping nuclear operators with specific solutions to fulfil audit criteria. The document also contains support information for NMAC auditors on the way of collecting information to assess the compliance with the audit criteria.

3.3 The audit procedure

The procedure to lead the involved actors in the process of audit has been drafted. This procedure is based upon the standard ISO 19011:2002 [3]**.** The main steps of the audit process as described in the procedure are:

- Initiation of the audit
- Document review
- Pre-audit, if necessary
- Preparation of on-site audit activities
- On-site audit activities
- Reporting

The procedure ensures that the timing of the audit activities is accepted by all the actors including the audit team and the nuclear operator. It also makes sure that audits will take place only when all the information needed is put at the disposal of the audit team. NMAC audit is seen by the European Commission as an activity adding value for the two organizations involved, namely, the nuclear operator and the nuclear inspectorate. NMAC audits have to be carried out in a collaborative environment.

The audit is initiated by the audit client, normally within the European Commission Directorate for safeguards the concerned Head of Inspection Unit, who nominates the audit team leader. After defining the scope and objectives of the audit, a feasibility study is performed in order to confirm that the resources for the audit are available and the information to

be collected will be at the disposal of the audit team. After nomination of the audit team members, the nuclear operator is contacted to agree on the scope, objectives, and a preliminary schedule for the audit. The documentation considered to be reviewed in advance is also requested at this moment.

A document review takes place with the objective of getting a global understanding of the operator's NMAC system and to detect differences between this system and the model described in the audit criteria. In case the document review can not take place in Luxembourg a pre-audit visit to the nuclear facility is foreseen.

A plan for the on-site audit activities is set where the tasks for each member of the audit team are detailed and the working papers are prepared.

The on-site audit activities start with an opening meeting, then the information gathering activities take place, and a closing meeting is held where the major findings and audit conclusions are communicated to the auditee.

The report is drafted by the audit team leader and after review and feedback from the audit client and eventually from the auditee, it is submitted for approval and distributed.

It is to note that the conduct of NMAC audits is not part of any certification process, in opposition to the audits performed to assess conformity to the most widely known quality management standards.

3.4 The audit approach

The use of NMAC audits is only justified when it ensures an improvement in the efficiency of nuclear safeguards processes. It is therefore not intended to use NMAC audits in a systematic way for all types of installations. The European Commission has identified five cases when NMAC audits can be used.

(1) *To assess the measurement systems of bulk handling nuclear facilities.*
Nuclear safeguards in bulk handling facilities are largely based on measurements done by the operator. Therefore, the efficiency of safeguards depends strongly on the quality and reliability of the operator's measurement systems. The Commission Regulation (Euratom) no 302/2005 of 8 February 2005 on the application of Euratom safeguards [9] states that the measurement systems of nuclear operators shall comply with the most recent standards or be equivalent in quality to those. Audit has shown to be an efficient and effective way to assess the quality of measurement systems. Accreditation is the internationally recognised method to guarantee the quality and reliability of measurement systems. Accreditation is based on the audit of the measurement systems to be accredited.

The most recent international standards about quality of measurement systems are ISO/IEC 17025:2005 [6] and ISO 10012:2003 [7]. The requirements stated in these two standards are faithfully reproduced in Section 4 of the Commission Recommendation (2009/120/Euratom) [4].

(2) *When a shortfall has been found in the operator's NMAC system.*
When a systematic weakness is found in an operator's NMAC system, an audit will find how this system can be improved, and so operator and inspectorate can react accordingly. Weak NMAC systems, reducing the efficiency of safeguards processes will increase the safeguards burden for nuclear operators and the effort of the inspectorate.

(3) *For installations joining the Euratom safeguards regime.*
A high quality NMAC system run by nuclear operators makes nuclear safeguards much more efficient. In order to ensure that nuclear operators run a high quality NMAC system when they join the Euratom safeguards regime, a NMAC audit may be carried out. Installations joining the Euratom safeguards regime include new built installations in the European Union or installations in countries acceding to the European Union.

(4) *For installations where the physical verification can be carried out only in a limited way.*
In some types of installations, the nuclear material is in such a form (or contained in such a way) that makes it by nature very difficult to perform physical verifications. Therefore, in

these installations safeguards rely on records produced by the nuclear operator. It is reasonable to assess, by means of audits, whether these records are produced according to proper procedures, and whether these procedures are checked appropriately. Typical examples of this case are waste handling and storage facilities.

(5) When a nuclear installation asks voluntarily for an audit of its NMAC system.

4. Training

In order to provide the European Commission with adequately qualified staff capable to perform the NMAC audit activities, a number of training courses covering audit techniques, NMAC systems, quality management systems and metrology are routinely taking place. However, the main source of qualification for the NMAC audit activities remains the experience gained on the job.

5. Conclusion

The European Commission has developed a sound framework for the use of NMAC audits for nuclear safeguards purposes. This framework is partly based on the outcome of the two ESARDA Working Groups that have been devoted to work on NMAC audits. The model used to develop the framework elements has been built upon the most widely recognised international standards.

After the development of the elements of the framework, the implementation of NMAC audits into the Euratom safeguards regime is taking place progressively. A number of audits have taken place during 2009, 2010 and 2011. The European Commission is conducting audits according to the latest international standard for audit services providers, namely EN ISO/IEC 17021:2006 Conformity assessment – Requirements for bodies providing audit and certification of management systems [10]. This standard contains structural requirements, resource requirements, process requirements and management system requirements for audit providers. The principles to follow in the conduct of audits are impartiality, competence, responsibility, openness, confidentiality and responsiveness to complaints.

The results of the NMAC audits that have taken place according to the framework described in this article show that safeguards can be enhanced by means of this tool.

6. References

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VARO - A Euratom software project for safeguards data evaluation

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Abstract:

Nuclear Safeguards implementation is more and more relying on electronic data and tools to evaluate these. The regular accountancy data being submitted to the Euratom Headquarters as part of the reporting requirements of the Euratom Regulation 302/2005 need to be checked against onsite data, operational records and compared with verification measurements. In modern, complex installations of the nuclear fuel cycle these data can easily amount to several thousand data sets, which need to be compared and evaluated for consistency.

In the past many existing safeguards applications have been tailor-made for specific installations, types of plants or even inspection teams and were mainly developed by experienced inspectors. This has led to a number of different safeguards applications in use, which over time have become more and more difficult to maintain.

With Euratom Regulation 302/2005 the required reporting format for nuclear operators in the European Union has changed from the 80 character line format to a xml based record set. These changes would have required an update of most of the existing applications to evaluate accountancy data, which proofed to be impossible, given the number of different applications and development platforms in question.

Since manual data treatment is not an option with the given time constraints during inspections, it was decided to start with the development of a software application for the evaluation and verification of accountancy data of operators, their comparison with operating records and related safeguards verification measurements.

The VARO (Validation of Accountancy Records of Operators) application aims to make safeguards evaluations and verifications to be coherent without limiting inspectors in their inspection scope. A generic application, centrally managed reduces resources needed for development, maintenance and training and will lead to a more consistent evaluation of safeguards data.

The application is supposed to become part of the standard inspection software package used by Euratom inspectors.

Keywords: data evaluation, accountancy, inspection support, software development

1. Introduction

All nuclear operators in the European Union are required to maintain a system of accountancy and control for nuclear materials. According to EURATOM Regulation 302/2005, these systems shall include accounting and operating records with information on quantities, category, form, composition and location of these materials, together with information on recipients or shippers, when nuclear materials are transferred.

Whereas the amount of these data might be limited for small installations and related safeguards verifications could be done manually, this has proven to be very resource demanding or even impossible in bigger or more complex installations. It is therefore essential to provide the inspectors with a dedicated software tool allowing them to carry out their verifications in an efficient and coherent manner.

2. Generic tools required

Almost immediately when computing power became more widely available, nuclear safeguards inspectors started to use this technology to evaluate nuclear accountancy data and related operational records. From simple spreadsheet type evaluations these developments evolved in a number of cases into very specific and complex applications that are essential for the successful execution of inspections.

However, the constant development of the hard- and software platforms, combined with staff mobility and the evolution of nuclear installations, have made it very difficult to maintain the numerous applications to keep them in working order and adapted to the inspection needs.

Apart from the difficult maintenance and long-term development these different applications created a constant need for inspectors training. When being on inspection inspectors are facing all sorts of difficulties and the number of different software applications in use for similar tasks is a further complicating factor that needs to be avoided.

In addition to these difficulties, the relevant reporting format for accountancy declarations was changed from 80 character lines to XML records with EURATOM Regulation 302/2005 coming into force, thus making most of the existing applications obsolete, because of the incompatibility of the input format.

The EURATOM VARO software project was started to address these difficulties and to develop a common verification tool that nuclear safeguards inspectors can use at different installation types and at headquarters to verify accountancy data against operational records and safeguards measurements performed.

3. Concept

EURATOM Safeguards uses database systems at their Luxembourg Headquarters (HQ) to process, evaluate and store the accountancy declarations that nuclear operators are required to send according to Regulation 302/2005 for most of the installations on a monthly basis. These records are first checked for compatibility of the report format, checked for internal consistency and then loaded into the accountancy database. The records are then checked against the reporting rules of the Regulation and facility specific criteria stored in a so called Rulebook database.

Nuclear safeguards inspectors carry out inspections to verify these declarations submitted by the operators with the physical reality at the installations. For this purpose they download from the HQ accountancy database relevant data for the planned inspection, including the last accountancy data sent by the operator. The deadline for transmission of the Inventory Change Reports (ICR) is the fifteenth of the following month.

The VARO application is designed as a web based application, which is running either on a HQ server or as a mobile version, using a so called Mobile Management System (MMS), which allows the creation of mobile kits used to export the different applications for use on inspections, including the relevant data to a storage media. Inspectors therefore always travel with the latest software version

and up to date data. The look and feel of the mobile version for VARO is exactly the same as with the HQ version. With the development of the Mobile Management System it is now possible to control the software development centrally, thus ensuring that the infield version is fully compatible with HQ and that data are handled in a coherent way.

Mobile Kit

All new safeguards applications at EURATOM Safeguards are now based on web interfaces, using Oracle databases. With the start of the development of VARO it was decided to develop a mobile working environment for essential safeguards applications to run on, called "Mobile Kit". Besides VARO applications for seal management (ESAM) and reporting (IMIS) have or will have mobile versions, thus allowing the inspectors to use their HQ tools when being out of the office. Since the sensitivity of safeguards data does not allow to use fully web based applications via internet interfaces and the EURATOM network is not accessible from the outside due to security considerations, these mobile versions work locally in an inspection context and are run either on the inspectors' notebooks, which have encrypted hard drives and are cleared to be used for the storage, treatment and transfer of these data. Before leaving for a mission the inspectors define the content of their specific mobile kits by selecting the required applications, the inspections to be covered and the data to be copied from HQ systems. Once the selections are made the mobile kit is prepared as a self extracting Java jar file serving as data and application container. The transfer of theses files is made on the encrypted hard drives of inspectors' notebooks or any other approved storage medium for sensitive safeguards data, like encrypted USB sticks with biometric user identification. The jar files can be opened directly on the inspectors' notebooks or at bigger installations, with safeguards IT infrastructure, on a server, thus allowing multi user access. In this respect the web based applications are versatile as they are multi user capable by design.

At the end of an inspection the preparation of an inspection outcome kit is necessary for the return of data to HQ. At HQ that outcome kit is used to upload the inspection data to the VARO HQ database. It is to be noted that there is no upload or synchronisation of accountancy data gathered during inspections with the HQ accountancy database. All data are stored in the inspection context for later reference, but not synchronised with other HQ applications. This was done to avoid any problem with data integrity and to keep the HQ data as reference.

Leaving for an inspection

The file management is based on an inspection context basis. Inspectors leaving for missions are able to carry in the mobile kits data for several different inspections. After having selected the relevant inspections, the inspector can change the predefined periods for HQ data copied into the Mobile Kit. Once the inspection data set is defined, the inspector can start with the Mobile Kit preparation. To protect the data a mobile kit password needs to be defined for the preparation and later installation of the mobile kit, which is independent of the password at HQ.

Depending on the inspection and installation either local PCs or inspector notebooks are used to run VARO. The mobile kits are self extracting Java jar-files that install the application in a directory defined by the inspector. The inspector notebooks use encrypted hard drives to ensure the required data security.

4. Working at the installation

On arrival at the installation the inspector either uses his inspector's laptop or at bigger installations the local PC network to install the mobile kit and run the applications. Having the same look and feel as with HQ applications should reduce the need for installation specific training and help inspectors' mobility. Having the relevant applications with them allows the inspectors work with data in near real time, thus limiting the risk of erroneous entries or reporting delays.

Accountancy data

One of the first checks during an inspection is if inventory changes happened since the last declaration that was sent to HQ. To do so, the operator is asked to provide an update of the accountancy data to the date of the inspection. These data are then loaded into VARO, checked for errors and can be merged with the HQ data contained in the Mobile Kit to provide the inspector with a seamless accountancy history up to the inspection date.

The accountancy data sent to EURATOM have to be in the format as defined in Regulation 302/2005. Many operators use the ENMAS application as provided by EURATOM, whereas others use plant, site or company specific applications to prepare these declarations. Since the required format is defined in detail, there is no need for plant specific data loading routines.

It is essential that inspectors use coherent verification rules when loading accountancy data. These rules are stored in HQ databases and are loaded into the mobile kits when preparing for an inspection. So there is only one reference, which is the HQ database, used for verification rules and the inspectors are sure that they are using the correct, up to date syntax for these checks as used in HQ and by the accountancy unit. After loading of the accountancy declarations, errors and warnings can be checked and the inspector has the option to compare and combine the accountancy declarations. Different browse, sort and filter options allow to adapt views and help the inspectors with their consistency checks. The resulting queries and settings can be saved by the inspectors, shared among different users and may be part of the MBA specific settings defined by the inspector in charge of the installation. This allows for consistent checks and routines to be used by all inspectors and avoids repeated definition of similar settings and rules. Print options allow preparing the different working papers and exporting to different file formats, e.g. txt, xls, pdf, which allow for using the data in other applications.

Operating data

Operating data are very plant specific and their detail and structure depend on the plant type, operational concept and size. Whereas in small installations the material tracking is done mainly manually using paper documentation, more complex installations with higher throughputs use IT systems that allow for online tracking of material flows and quantities in the different areas. This serves many purposes, like process control, quality assurance, criticality control, product documentation and is also important for nuclear material accountancy and control. Plants using fully integrated material tracking systems normally have very good documentation of their operations and are able to provide this information in electronic format to the safeguards authorities.

The consistency between accountancy and operating data is one of the essential checks inspectors carry out during their verifications. The VARO application is supposed to help inspectors with the evaluation of operating data and their comparison with accountancy declarations. However, since operational data and their structure are plant specific it is essential to normalise them to an internal data structure that is compatible among all installations. To load installation specific data with different data structures a mapping between these and the internal database is required. For this purpose data loaders need to be defined, that translate the different formats into the internal structure.

The standardised internal data structure does also allow using common queries that can be shared amongst groups of inspectors and installations, as it is done for the accountancy data.

The matching of operating with accountancy data should normally be possible in a semi automatic way. The application will check the operating data and try to find matching accountancy lines. The inspector has always the option to accept these matches or to match them manually. To help the inspectors with the matching, detailed query functions exist as evaluation tools. As for accountancy data, report and export functions allow for documentation and further checks using other applications.

Document checks and versioning

During the course of an inspection it is sometimes necessary for the operator to update documentation provided. Since inspectors often immediately process information made available, each updated set of data causes a problem for the inspectors to know which data were changed and if these changes have an impact on the verifications already performed or the conclusions already drawn. A typical example is the List of Inventory Items, which as an operational document, is sometimes updated by the operator during a PIV. It is therefore essential to have a routine to check updated data files against the original and to highlight changes found. This avoids double checks, ensures at the same time that all items are verified or have a chance to be selected and also allows for a statistical evaluation on the quality of original documents as provided by the operators' NMAC systems.

Measurement data

Physical verifications are the most essential part of inspection activities. After the accountancy data have been verified against operating records, these are used to sort the nuclear material into different

strata depending on the material category, location and available instrumentation. Depending on the inspection type, lists of inventory items (LII), store inventory listings or movements at strategic points of the plant are used to select items for measurements. The selections are made using a sampling algorithm, which is depending on the required detection probability, detection goal quantity and average material quantity of the strata. These requirements are part of the MBA specific set up, which is pre-defined by the inspector in charge at HQ, thus ensuring that all inspectors use the same criteria when planning their physical verifications. These requirements are stored in installation specific queries, which are also part of the mobile version of VARO that travels with the inspectors.

Once the item selection for measurements is done, the inspectors prepare print outs for use as working papers in the installation, or use the relevant data entry screen of the VARO application directly to record their verification results.

After having carried out the physical verifications the inspectors have to enter the measurement data into the application, either manually or using instrument specific import routines.

Instrumentation running in unattended mode is often used to monitor process flows at strategic points in the installations. These instruments are controlled with specific software and the resulting measurement data are stored at the local data acquisition modules (DAM) and automatically transferred to data servers. Evaluation routines are used to find the relevant measurements and to combine related signals, e.g. neutron and gamma data or weighing results with bar code readers, to measurement events.

By linking these data with the operating data a direct comparison and statistical analysis becomes possible. The linking of measurement events with operating data is normally done automatically by the VARO application. However, the inspector is able to overwrite these links in case of a wrong matching.

By running automatic comparisons between the two data sets, the inspectors get immediate feedback, if measured results are outside of the acceptance criteria and have the option to recheck or re-measure. At the end of the physical verifications specific routines and listings allow the inspectors to calculate overall statistics and to document their results.

Data transfer to HQ

Normally the inspectors enter most of the relevant data into the VARO application onsite and have them available for later use, evaluation, documentation and statistical analysis. However, some Member States have very specific legal restrictions on the transfer of data related to nuclear material quantities and whereabouts. To address these restrictions it is possible to limit the data travelling back with the inspectors to HQ by taking only pre-defined data sets back, i.e. without location information, with them.

5. Return to HQ

On return to HQ the inspectors are required to upload their inspection data to the HQ database. After this upload it is possible to continue with the data evaluation seamlessly. The use of the mobile version allows for the inspectors' independence on onsite data and takes away the pressure to finish evaluations onsite.

Dedicated data output formats and print outs allow the to document their inspection activities and verification results, which are used as annexes to the inspection reports, which are the overall documentation and evaluation of the inspection activities.

After the evaluations are finished it is the responsibility of the Head of the Inspection to close the data file in VARO. This is the moment when the inspection data are fixed in the HQ database and considered as reference.

Data storage

Whereas the HQ database for accountancy data is to be seen as reference, similar central repositories for operational and measurement data do not exist. These data are normally stored at the

installations and taken, if possible, back to HQ as separate files. These files have different structures, depending on their origin and are not stored in a single, compatible data format.

With the start of the VARO project it was decided to normalise operating and measurement data using an internal data structure that allows for a central storage.

However, depending on the installation type, it might be necessary for the inspectors to reference back to data received earlier or to have older measurement data available for comparison and to complement data. This is for example necessary at storage locations used at fabrication facilities, where isotopic composition details are needed for later in-process measurements. Since the VARO concept is relying HQ data as reference it is essential, that inspectors are able to take relevant operating and measurement data for predefined areas and periods with them when leaving for an inspection.

Statistical evaluation

The centralised HQ storage of relevant inspection data allows not only for a statistical evaluation in the inspection context, but also for data analysis in a wider context. It is foreseen to have statistical routines allowing for periodical evaluations on MBA or installation level. This will help the inspectorate to better focus the inspection effort and to adapt it regularly to requirements.

6. Present status of the development

The development is split into different phases to allow for a phased introduction and to avoid developments that prove to be inadequate for prolonged periods of time. Phase one of the project was focused on the accountancy evaluation. This part of the application is needed at all installations and addresses the most urgent need of the inspectors. This part of the application is already in use and first development iterations, taking into account shortcomings found and other user comments, have been implemented. Phase 2 of the project is focused on the loading and evaluation of operating data and their comparison with the accountancy data. This part is under development at the moment. Phase 3 is foreseen to address the stratification, sampling and comparison of measurements with the operating data. Additional phases are foreseen for the statistical evaluations, user specific settings and their storage.

7. Co-operation of all stakeholders required

The scope of the VARO application is besides other elements an increased efficiency of the inspection resources available. It must be in the interest of all stakeholders to carry out inspections as smooth as possible and without any unnecessary delays. One element in this equation is an efficient and effective data treatment. Whereas in almost all installations the use of computerised material tracking systems is established practice, the data exchange with the safeguards authorities is often done in a patchwork fashion. To avoid cumbersome and time consuming manual data treatment during inspections, it is essential that inspectors receive predefined datasets, which can be loaded and evaluated automatically. To support the use of VARO inspectors are required to agree common data interfaces with the operators. This will save time and effort at all sides, but requires an initial investment and co-operation from operators and national authorities to get these arrangements into routine operation. Another main player is the IAEA, which is informed about the project and have been invited to indicate further specific requirements.

Recent Developments in Safeguards Practices in the European Union

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Abstract:

The IAEA applies Integrated Safeguards in 24 non-nuclear-weapon states (NNWS) of the European Union since early 2010. The nuclear safeguards regime in these States is based on the Euratom Treaty combined with a trilateral comprehensive safeguards agreement (INFCIRC/193) and its Additional Protocol (AP) between the IAEA, the Member States and the European Atomic Energy Community (Euratom). Since the entry into force of the AP in 2004 the IAEA combines new safeguards tools for assuring the absence of undeclared nuclear material and activities with the traditional ones.

The European Union countries possess a complete nuclear fuel cycle (NFC), with hundreds of civil installations, from large to very small, all subject to the control of Euratom, the regional system of accounting for and control of nuclear material (RSAC). While new nuclear power production capacity is being planned and built in the Union, questions related to the decommissioning of former installations and the management of waste are topical in many parts, due to the early start and/or subsequent change of priorities in favour of other activities.

This paper takes a look into the process that led to the broader safeguards conclusions for the EU States and the introduction of Integrated Safeguards. Apart from providing an overall view of the key issues, it discusses specific elements from the perspective of in how far the strengthened tools (information, access, activities) have become an integral part of safeguards practice and are influencing the use of traditional safeguards measures.

Keywords: Implementation; Strengthened Safeguards; Integrated Safeguards; implementation

1. Nuclear Safeguards System in the European Union

The system of nuclear safeguards in the European Union was established in Chapter 7 of the Euratom Treaty [1]. Article 77(a) of the Treaty makes the European Commission responsible for ensuring by appropriate supervision that nuclear materials are not diverted from their intended uses. Article 77(b) requests that the European Commission makes sure that obligations undertaken under Agreements with third States or international organisations are fulfilled. The Safeguards Agreements between the Member States, Euratom and the IAEA, and their corresponding Additional Protocols belong to these agreements [2]. The Commission Regulation on the application of Euratom safeguards further specifies the provision of the Euratom Treaty and the Safeguards Agreements, including the Additional Protocol, for the holders of nuclear material in the Union [3]. What follows in this paper concerns mainly the Safeguards Agreement between the IAEA, the European Atomic Energy Community and the NNWS, INFCIRC/193.

Recognising the Commission's role as the safeguards authority of the European Union (EU), the Safeguards Agreement requires the IAEA to make full use of the Euratom system of safeguards and avoid unnecessary duplication of safeguards activities. These requirements are further specified in the Protocol to the Agreement.

The Protocol also lays down the establishment of a body consisting of members assigned by the two authorities for facilitating and monitoring cooperation. This body, the Liaison Committee, is required to

meet at regular intervals. In practice, a High-Level Liaison Committee (HLLC) meets approximately annually, a Low Level Liaison Committee (LLLC) semiannually, while dedicated working groups discuss technical questions and report to the LLLC. Close cooperation and coordination is instrumental for ensuring that both organisations can reach their safeguards goals and make independent conclusions based on activities carried out in joint inspections, using common equipment.

2. Towards Integrated Safeguards in the European Union

The entry into force of the Additional Protocol (AP) to INFCIRC/193 (INFCIRC/193/Add.8) can be seen as the starting point of the last *étape* in the journey towards the introduction of Integrated Safeguards (IS) in the European Union. Five years and eight months passed from the date of that event (30 April 2004) to the date of 1 January 2010 when the IAEA was ready to start implementing its IS regime in the 24 NNWS of the EU's 27 Member States.

During these preparatory years many changes took place. The EU was enlarged by 10 new Member States on 1 May 2004 and by two more on 1 January 2007. They all became subject to the Euratom Treaty safeguards system immediately on joining the EU, but the process for adopting the common international safeguards agreement INFCIRC/193 took time. During this interim period, the bilateral safeguards agreements that the States had earlier made with the IAEA remained applicable. The transition was completed State by State, starting on 1 December 2005 (Estonia and Slovakia) and ending with the accession of Romania on 1 May 2010. Since then the legal basis to apply safeguards in the EU is homogenous again.

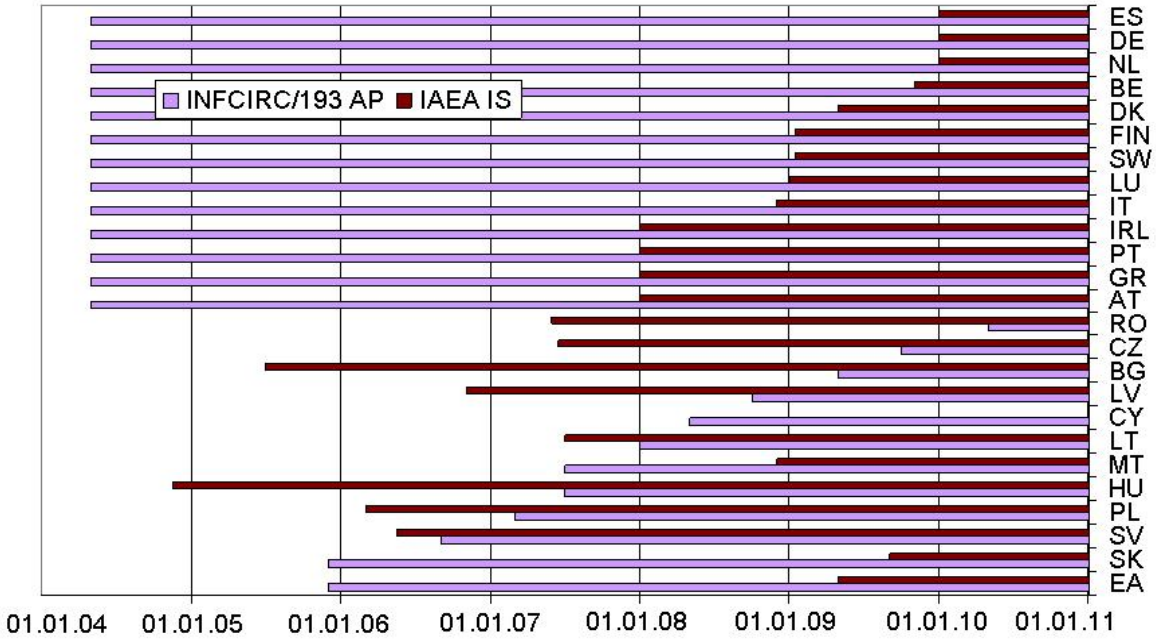

Chart: Implementation of INFCIRC/193/Add.8 and of Integrated Safeguards

As regards the AP to INFCIRC/193, INFCIRC/263 and INFCIRC/290 the initial declaration for the 15 EU Member States concerned was made at the end of October 2004. The implementation of Complementary Access started in December 2004. Over the following year the IAEA requested a large number of amplifications and clarifications to the information provided. The new Member States having joined the EU after the entry into force of INFCIRC/193/Add.8 were required to either join the AP for the first time (Estonia and Slovakia) or switch from the AP to their bilateral safeguards agreements to that of the INFCIRC/193 on accession to the common safeguards agreement.

The switch was accompanied by the provision of an 'initial declaration' to INFCIRC/193/Add.8. As the content of the AP remained the same as before, apart from the attribution of tasks, no major problems

arose. The Commission applies harmonised standards to the presentation and details in the content of the declarations. This led in some cases to a slightly different declaration content than under the bilateral AP's. While not putting the Commission's approach into question, some expressions of discontent because of 'changed' content were heard from the Agency. In order to facilitate the IAEA's implementation of the AP, it was agreed for these new Member States that, during the time from the entry into force of the common AP until the initial declaration pursuant to INFCIRC/193/Add.8 was provided, the IAEA could continue its AP activities (CA) on the basis of the bilateral declarations.

The IAEA had drawn the first broader conclusions for most of the Member States having acceded to the EU in 2004 already before their accession to INFCIRC/193. The introduction of Integrated Safeguards took place in Hungary still in 2004 and in other States in the following years. The IAEA thereby also introduced its new safeguards practices in these countries before their accession to INFCIRC/193.

In parallel with the new Member States moving from the status 'bilateral safeguards agreement without IS' via 'bilateral safeguards agreement with IS' to 'INFCIRC/193 with IS', the 'old' Member States proceeded towards receiving their first broader safeguards conclusions and the approaches for the implementation of IS under INFCIRC/193 and its AP.

In terms of the implementation of the AP, the IAEA devoted considerable effort to verifications of what could be related to not only on-going but also past undeclared nuclear activities. A large number of requests for amplification or clarification were made and CA was conducted across the EU, in different types of facilities. At the same time status verifications at former installations continued, very small holders (the CAM) were 'screened' and some of them inspected as a one-off voluntary measure because they are normally not subject to regular inspections.

In preparation for the time when all NNWS of the EU would be covered by the one and same IAEA safeguards framework – 'INFCIRC/193 with IS' – contacts were initiated within the Euratom-IAEA cooperation scheme, in order to agree on the necessary changes to ensure that both organisations could continue fulfilling their objectives. Discussions on the cooperation on the way to IS started in 2006 and continued mainly in the framework of the Liaison Committee meetings thereafter. The main lines for the future cooperation arrangement were agreed and the work on preparing the detail could start. Although the devil was also here in the detail, the arrangements could be finalised in time to enable each next step in the start of implementation of IS in the EU under a common umbrella.

3. The Integrated Safeguards Framework in the EU

The basic requirements for the new common safeguards framework in the EU were set by (i) the IAEA's requirements for the implementation of IS, and (ii) the need to ensure that the Commission as the European safeguards authority could fulfil its tasks under the Treaty in cooperation with the IAEA. The cooperation framework included key principles such as

- The principle of common or joint inspections ('one person for one job');
- Sharing of inspection documents in cases where one of the organisations was not present;
- Inspection planning done by the Commission;
- Common use of equipment.

In this respect it has to be noted that the HLLC confirmed that the general cooperation principles that were laid down in the so-called New Partnership Approach (NPA) of 1992 would continue to be valid.

The IAEA's generic approaches for Integrated Safeguards contained requirements for new features to be incorporated into the common framework. These included the use of unpredictability, including the randomization of inspections, combined with very short or no advance notice time, in order to reduce inspection effort. While Euratom also saw potential benefits in the concept of making some inspections unpredictable to the operator, some aspects of the implementation proposed by the IAEA were problematic to the European safeguards authority. Without an early notification by the IAEA to the Commission of a short-notice random inspection (SNRI) or an unannounced inspection (UI), it would be very difficult (SNRI) and impossible (UI) to attend.

As a result of the negotiations, the use of UI was restricted to a limited number of installations (currently 10), where the installation of adequate surveillance or the use of other technical means to avoid UI either is not possible or has not yet been made. In line with its safeguards approach, Euratom will continue to use surveillance and equip installations with surveillance where feasible. The use of surveillance creates the necessary conditions for an inspection regime where interim inspections can be performed jointly. In addition, surveillance offers to the IAEA a valuable back-up measure. For the SNRI's – currently around 50 per year in more than 100 installations in 14 Member States – a system of 'early indication' was elaborated. It combines the IAEA's request to ensure the surprise effect towards the operator with the Euratom need to receive an early notice, in order to have an inspector available and able to reach the installation within the short (24- or 48-hour) advance notice period. A three-month rolling plan is updated every month by the IAEA, listing for each week the number of planned SNRI's, however, without any indication of the location. Apart from scheduling the SNRI's and UI's, it was agreed that the IAEA would also schedule and trigger a very limited number of unannounced accesses (LFUA) in enrichment plants.

The arrangements for the different types of installations were recorded in Partnership Approach (PA) papers, which were modelled on the NPA papers that had set out detailed implementation arrangements between the IAEA and Euratom since the early 1990's. Generic papers were prepared for the main installation types: light-water reactors (LWR), research reactors (RR), spent fuel storage facilities (SFSF), fuel fabrication plants (FFP) and gas centrifuge enrichment plants (GCEP). Where necessary, an installation specific PA paper complements the generic one, setting out specific provisions for that installation (mainly FFP and GCEP). Specific papers for the two on-load reactor facilities in the EU are currently under preparation. An important feature, also for the operator, in the context of IS is the requirement to provide more frequent updates of activity programs. The daily or weekly provision of updated production programs, together with the need for product or feed retention time (availability for inspection), were amongst the much discussed topics in relation to FFP's and GCEP's.

A series of PA papers for inspection support were also prepared. These concern matters such as arrangements for joint-use remote data transmission (RDT); the sealing arrangements for joint use equipment; and spent fuel cask loading and sealing.

Facility-specific lists of inspection activities were prepared on operators' request for some larger installations, in order to help the operator prepare for SNRI's. These papers have been appreciated by the operators, who had initially feared not to be able to fulfil the expectations put on them by the new short-notice regime.

4. Implementation Experience

4.1. Inspection Effort

The IS paradigm permits the IAEA to reduce the verifications of declared nuclear material, in order to focus also on ensuring the absence of undeclared nuclear material and activities. The application of unpredictability in routine inspections has allowed the IAEA to devote more resources to activities linked to the search for undeclared material and activities. Fewer inspection days means smaller inspection effort to the IAEA.

The reduction has been largest in the LWR's, as both the IAEA and Euratom have adopted the scheme relying on a small number of SNRI's as interim inspections. In other types of installations the savings in inspection days have been smaller and so far mainly achieved by the IAEA, as Euratom has not reduced its presence in the installations. This is particularly true for fuel fabrication plants (FFP) and enrichment facilities.

The other side of the coin in randomizing inspection activities is the additional charge that the change may cause to the nuclear operators. The gain in the reduced inspection days may be outweighed by the requirements for more frequent information updates or short preparation times. Whether the price to pay for the confirmation that no diversion of nuclear material has taken place, has decreased may depend on the installation.

The strict application of the principles of randomisation has caused astonishment on a few occasions, as an SNRI has been announced almost immediately before or after a regularly planned inspection in the same installation.

4.2. Diverging Inspection Schemes

While both the IAEA and Euratom can reach their respective safeguards goals under the current implementation schemes, the somewhat diverging schemes may lead to less than optimal implementation. In their current approaches the IAEA emphasise strongly randomisation and the benefits derived from it. Euratom has also embarked to this path, but in a more limited manner. Random approach is currently in use for light water reactors (LWR) in the NNWS, allowing to carry out all inspections jointly in these installations. In order to avoid divergence of approaches across the Member States of the EU, Euratom, in line with its inspection guidelines, is introducing short-notice or unannounced inspections to the power reactors in the United Kingdom and France, where applicable.

A scheme of random inspections is also used in the fuel fabrication plants where both Euratom and the IAEA trigger a certain number of such inspections.

The coordination of inspection activities between the IAEA and Euratom for GCEP's is achieved in a framework whereby Euratom plans and carries out a larger number of inspections than the IAEA, which allows the IAEA to participate in the required number of Euratom inspections. Although this arrangement aims at avoiding all duplication of inspections, there has been a tendency to duplicate some activities. This has happened in some occasions where the IAEA, not having been present during the previous inspection, requested that activities already carried out be repeated.

In all cases where random inspections are used the system of a planned annual PIV obviously continues to apply.

Euratom considers that for reasons of keeping up their familiarity with smaller installations, annual or bi-annual inspections are needed. For these installations the IAEA has chosen a random system whereby each small installation should be selected for PIV inspection once in 4-6 years.

In the NNWS, a difference in the number of inspections carried out by Euratom inspectors alone and carried out together with inspectors of the IAEA should however not be considered as a divergence of the inspection schemes. For Euratom it is clear that, especially in a process which leads to the IAEA making more of use the Euratom safeguards system, there should be more inspections done by Euratom inspectors alone and the IAEA being provided with all information and results about these inspections. However, any divergence of the schemes in the direction of duplication of inspection activities or rejecting the use of information and results from inspections carried out by the other party should be opposed.

4.3. Notification times

The IAEA's requirement to limit the notification time for the SNRI to 24 hours applies in most installations where random inspections are in use. Apart from a few cases, Euratom also receives the notification only 24 hours in advance. In other cases Euratom is notified 48 hours and the operator 24 hours in advance. Despite the fact that, based on the rolling plan, an inspector has been already assigned to the task when the notification arrives, it has turned out that the time required by the installation-specific preparations together with the travel arrangements make it difficult to reach some locations within the shorter, 24-hour notification period, while ensuring a few hours of rest to the inspector between arrival and the start of inspection.

For the future it will be important to review the relevant parameters of the SNRI arrangement, in order to ensure safe travel and working conditions to the inspector. Euratom has made a request to extend the list of installations where the 48-hour notification period applies.

4.4. Remote Data Transmission

Euratom, in cooperation with the Joint Research Centre (JRC), has been studying and promoting the use of newest safeguards technologies, with the aim of improving efficiency and effectiveness by a

more rational split between work in the field and in headquarters. Remote data transmission is seen as offering sizeable savings in terms of inspection days in some types of installations. The progress has, however, been very gradual in some Member States, as fears related to technical features designed to ensure data security have emerged, while other more generic fears on operational privacy also remain to be resolved. In a number of installations, RDT is already in use, with the data being transmitted to both safeguards organisations. RDT is also an essential means for getting assurance that surveillance systems properly work in those installations where these are a condition for the use of SNRI.

In future enhanced use of RDT will help moving inspection activities from the field to the headquarters, which will further reduce the burden to the nuclear operators.

4.5. Strengthened Safeguards/AP

The AP opened many new doors to the IAEA by permitting access to a wide range of locations, without the need to separately agree on the timing and other modalities. Complementary Access is available for the purpose of obtaining assurance of the absence of undeclared nuclear material or activities, confirming the decommissioned status of a former nuclear installation or resolving a question or inconsistency in the information provided by the State or the Community. An additional requirement is that the IAEA will not mechanistically or systematically check declarations that have been provided. A request for access can target a declared location or an undeclared location. In the latter case the access must be preceded by an opportunity for the State or the Community to facilitate the clarification of the question. The Agency here has ample room for discretion to act.

By making a wide use of its CA rights the IAEA has made sure that these rights did not become a dead letter in the AP. Beyond the purpose of affirming its access rights and training its staff, the equal treatment of all countries has sometimes surfaced as a reason for making frequent requests for CA, mostly to declared locations in all Member States. Altogether 210 CA's were made in the past 6 ½ years in the States party to INFCIRC/193/Add.8. Taking into account the questions asked and other verifications made this experience gives a strong indication of CA being used in the EU as an additional tool for routine verification. To carry out the 44 CA's that took place in the EU in 2010, the IAEA spent over 200 person days in the field.

The IAEA has been putting strong emphasis on obtaining access within the minimum advance notice time stipulated in the AP. The intention of the AP was that the Agency use judgement in setting the CA parameters, including the advance notice time, depending on the technical objectives. In a few cases the short notice time has limited the access that could be provided. Arrangements for more access could have been made if the specific places (buildings) had been announced before the beginning of the CA. The IAEA is taking the conscious risk of getting less access rather than giving (longer) advance notice, i.e. setting the notice time based on the technical purpose of the access. Beyond objective difficulties, the short notice time tends to put a psychological pressure on the operator and the safeguards authority.

As regards CA activities, the swipe tests using HPTA/ES technique provide a powerful tool for tracing activities involving nuclear materials far to the past. Swipe tests have been widely applied also in the CA context in the EU. Practical factors have, however, somewhat complicated their use. CA swipes are mostly taken in places for which no baseline data exists. This makes the availability of complete and reliable information about the nuclear materials used and activities carried out in the location of swipe-taking decisive for the interpretation of the results and possible follow-up action. The collection of such information (the operator's statement) often proves difficult during CA. The operator's representatives may not be familiar with the complete nuclear history of the location; or they may not be aware of the use of the information being provided. It would be important to clearly explain the context, assess the quality of information received and ask specific questions. CA swipes have had the tendency to lead to a number of apparent 'discrepancies' and follow-up questions or activities.

The CA appears to have been implemented as an activity that is planned long in advance and kept rather separate from other safeguards activities. CA does not respond to recent information, and in only one or two cases has there been a timid attempt to apply CA for clarifying a safeguards-relevant event, such as the loss of surveillance. The CA request, however, came long after the triggering event.

5. Towards Further Integration of Safeguards

Based on the experience, there is way to go before IAEA safeguards can be called truly integrated. Flexibility in function of different situations in different States, as well as responsiveness to events challenge the safeguards organisation.

Flexibility and differentiation are key words, if the IAEA is to take full advantage of all information available to it. While a lot has been said about including 'state-specific factors' in evaluation, it is not clear how the Agency intends to do so. As an example of an essential state-specific factor, let us mention the concept of transparency. Its meaning and use would deserve to be developed, however, different from the interpretation that the Agency assigns to it today. Transparency should be assessed based on a systematic analysis of institutional and social structures that are prevailing in a State in respect of diversion scenarios. A sort of 'social model' could be developed and used in a way similar to the 'physical model' of diversion.

When considering State-specific factors it is more than obvious that a difference has to be made by the IAEA between States which have a single state SSAC and states that are covered by a regional system (RSAC). Further integration and savings could be achieved by enhancing the use of information, especially from RSAC's with adequate technical capability, such as Euratom. The inspection documents and results provided by Euratom from the inspections where the IAEA is not present could be immediately used. This, intelligently combined with CA where needed, would allow a further reduction of the Agency's inspection effort in Europe and the shift of resources to other regions of the world, where strong regional safeguards systems do not exist.

As the use of CA has now become established, the frequency should be reduced towards the real needs by making use of the flexibility that the AP permits. This relates to daring to treat different situations differently, be it between States, the locations targeted or the advance notice given. Rather than routine checking in X out of Y declared locations of type k, however not less than 1 in each State with nuclear activities each year, the basis for asking CA would be a concrete suspicion related to the target location. Written questions and clarification request would serve not only to obtain access to an undeclared location, but also to resolve doubts.

The integration of AP activities to 'classical' safeguards activities in the IAEA could lead to new uses for CA, thereby avoiding too dramatic decline in the numbers in the EU. CA could be employed e.g. for solving some situations of anomalies and verifying the status of certain closed-down facilities.

As regards Euratom, the integration would usefully start from the genuine inclusion, or embedding, of the AP into the safeguards organisation. Once this step is taken, the questions and answers related to the role of Euratom in the implementation would follow naturally. The reflection required would also allow taking a new look into the role of Euratom in the European safeguards.

6. References

[1] *Consolidated version of the Treaty establishing the European Atomic Energy Community, consolidated version; Official Journal of the European Union (OJ) C 84 of 30.3.2010*; p 1.

[2] INFCIRC/193: *Agreement between the Kingdom of Belgium, the Kingdom of Denmark, the Federal Republic of Germany, Ireland, the Italian Republic, the Grand Duchy of Luxembourg, the Kingdom of the Netherlands, the European Atomic Energy Community and the International Atomic Energy Agency in implementation of Article III (1) and (4) of the Treaty on the non-proliferation of nuclear weapons (78/164/Euratom);* OJ L 51, 22.2.1978, p. 1.
(All NNWS that have subsequently joined the EU have also acceded to INFCIRC/193 and its AP.)

INFCIRC/193/Add.8: *Additional Protocol to the Agreement between the Republic of Austria, the Kingdom of Belgium, the Kingdom of Denmark, the Republic of Finland, the Federal Republic of Germany, the Hellenic Republic, Ireland, the Italian Republic, the Grand Duchy of Luxembourg, the Kingdom of the Netherlands, the Portuguese Republic, the Kingdom of Spain, the Kingdom of*

Sweden, the European Atomic Energy Community and the International Atomic Energy Agency in implementation of Article III(1) and (4) of the Treaty on the Non-proliferation of Nuclear weapons (1999/188/Euratom); OJ L 67, 13.3.1999, p. 1.

INFCIRC/263: *Agreement between the United Kingdom of Great Britain and Northern Ireland, the European Atomic Energy Community and the International Atomic Energy Agency for the Application of Safeguards in the United Kingdom of Great Britain and Northern Ireland in Connection with the Treaty on the Non-Proliferation of Nuclear Weapons*

INFCIRC/263/Add.1: *Additional Protocol to INFCIRC/263*

INFCIRC/290: *Agreement between France; the European Atomic Energy Community and the International Atomic Energy Agency for the Application of Safeguards in France*

INFCIRC/290/Add.1: *Additional Protocol to INFCIRC/290*

[3] *Commission Regulation (Euratom) No 302/2005 of 8 February 2005 on the application of Euratom safeguards;* OJ L 54 of 28.2.2005, p. 1.

05 Advanced measurement techniques for Spent Fuel

LA-UR 11-02438

Use of Self-Interrogation Neutron Resonance Densitometry to Measure the Fissile Content in a BWR 9x9 Spent Fuel Assembly

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Abstract:

We have investigated the use of Self-Interrogation Neutron Resonance Densitometry (SINRD) to measure the fissile content in a BWR 9x9 spent fuel assembly in water via Monte Carlo N-Particle eXtended transport code simulations. In addition, the sensitivity and penetrability of SINRD to the removal of fuel pins from an assembly was also assessed. The sensitivity of this technique is based on using the same fissile materials in the fission chambers as are present in the fuel because the effect of resonance absorption lines in the transmitted flux is amplified by the corresponding (n,f) reaction peaks in fission chamber. These simulations utilize the ^{244}Cm spontaneous fission neutrons to self-interrogate the fuel pins. The amount of resonance absorption of these neutrons in the fuel can be measured using ^{235}U and ^{239}Pu fission chambers placed adjacent to the assembly. Ratios of different fission chambers were used to reduce the sensitivity of the measurements to extraneous material present in the fuel. SINRD requires calibration with a reference assembly of similar geometry. However, since this densitometry method uses ratios of different detectors, most systematic errors related to calibration and positioning cancel in the ratios. The development of SINRD to measure the fissile content in LWR spent fuel is important to the improvement of nuclear safeguards and material accountability. Future work includes performing experimental measurements with a prototype SINRD detector pod on both fresh and spent LWR fuel in water.

Keywords: spent fuel; nuclear safeguards; non-destructive assay; plutonium

1. Introduction

The development of non-destructive assay (NDA) capabilities to directly measure the fissile content in spent fuel is needed to improve the timely detection of the diversion of significant quantities of fissile material. This NDA capability is important to the implementation of integrated safeguards for spent fuel verification by the International Atomic Energy Agency (IAEA) and would improve deterrence of possible diversions by increasing the risk of early detection [1]. The IAEA requires effective NDA methods to verify the spent fuel and recover continuity of knowledge in the event of a containment and surveillance systems failure [2]. Furthermore, this assay capability would also improve material accountability information at reprocessing plants prior to fuel dissolution and thus increase operational efficiency, reduce material unaccounted for (MUF), and reduce shipper-receiver differences (SRD) [3].

The primary objective of this research is to develop and assess the sensitivity of using Self-Interrogation Neutron Resonance Densitometry (SINRD) for nuclear safeguards measurements. Recent interest in this approach was stimulated by an IAEA request related to spent fuel verification. Prior measurements [4,5,6] and calculations [7] have demonstrated that the SINRD method gives quantitative results for the fissile concentration in metal plates, MOX fuel rods, and a PWR 17x17 fresh fuel assembly [8].The main application of SINRD is for use at a spent fuel storage facility for measurements in water, although SINRD could also be used for measurements in different mediums, such as air or sodium, and at reprocessing facilities that have spent fuel pools. The focus of the work described in this paper was to investigate the viability of using SINRD to verify a BWR 9x9 spent LEU fuel assembly (FA) via Monte Carlo N-Particle eXtended transport code (MCNPX) [9] simulations. The

following capabilities of SINRD were assessed: 1) ability to measure the ^{235}U and ^{239}Pu content in BWR spent LEU fuel and 2) sensitivity and penetrability to the removal of fuel pins from an assembly.

The neutron resonance cross-section structure is unique for different fissile isotopes such as ^{235}U, ^{239}Pu, and ^{241}Pu. This resonance structure can provide a signature for the measurement of materials of importance to safeguards and non-proliferation. The sensitivity of SINRD is based on using the same fissile materials in the sample and fission chamber because the effect of resonance absorption in the transmitted flux is amplified by the corresponding (*n,f*) reaction peaks in the fission chamber. For instance, a ^{235}U fission chamber has a high sensitivity to the neutron resonance absorption in ^{235}U present in the sample, and similarly for other fissile isotopes [4,8,10].

In Figure 1, the ^{239}Pu fission cross-section is compared to the resonance absorption lines in the neutron flux after transmission through a 0.11-mm Gd filter and ^{239}Pu metal samples 0.25-mm and 2.5-mm thick [4]. It is important to note that as the sample thickness increases, the self-interrogation signature also increases which decreases the transmitted flux reaching the FCs. Thus, the self-interrogation signature is inversely proportional to the amount of resonance absorption in the sample. The results shown for the transmitted flux through ^{239}Pu metal samples of different thicknesses were obtained from MCNPX simulations and the ^{239}Pu fission cross-section was obtained from the JANIS ENDF-VII cross-section database [11].

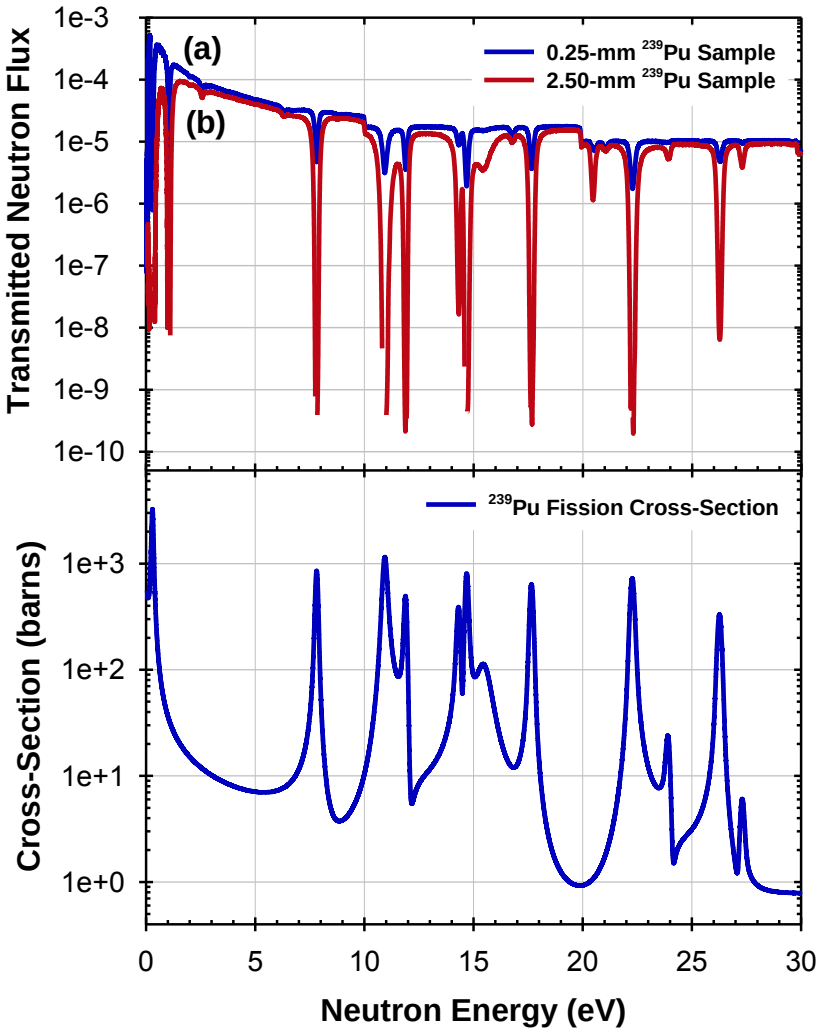


Figure 1. Comparison of absorption lines in neutron flux after transmission through Gd filter and (a) 0.25-mm and (b) 2.5-mm ^{239}Pu metal sample (upper plot) to ^{239}Pu fission cross-section (bottom plot).

2. Description of SINRD Measurement System

We have simulated the use of SINRD to quantify ^{235}U and ^{239}Pu in spent fuel and detect possible diversion scenarios for a BWR 9x9 spent LEU fuel assembly in water with 0%, 40%, and 70% void fractions (VF). This required first calculating the isotopic composition of the spent fuel assemblies using TransLAT [12] over burnup range of 0 to 50-GWd/MTU (in 10-GWd increments). Then, SINRD's response to each assembly was simulated. The concentration of ^{235}U and ^{239}Pu in the spent fuel pins was determined by tallying the fission rate in ^{235}U and ^{239}Pu fission chambers (FCs) located adjacent to the fuel assembly. Spontaneous fission neutrons from ^{244}Cm were used to self-interrogate the spent fuel pins in the MCNPX simulations of SINRD. It is important to note that the spent fuel isotopics were assumed to be homogeneously distributed in the fuel pins in the simulations. The specifications used to model a BWR 9x9 spent LEU fuel assembly are given in Table 1.

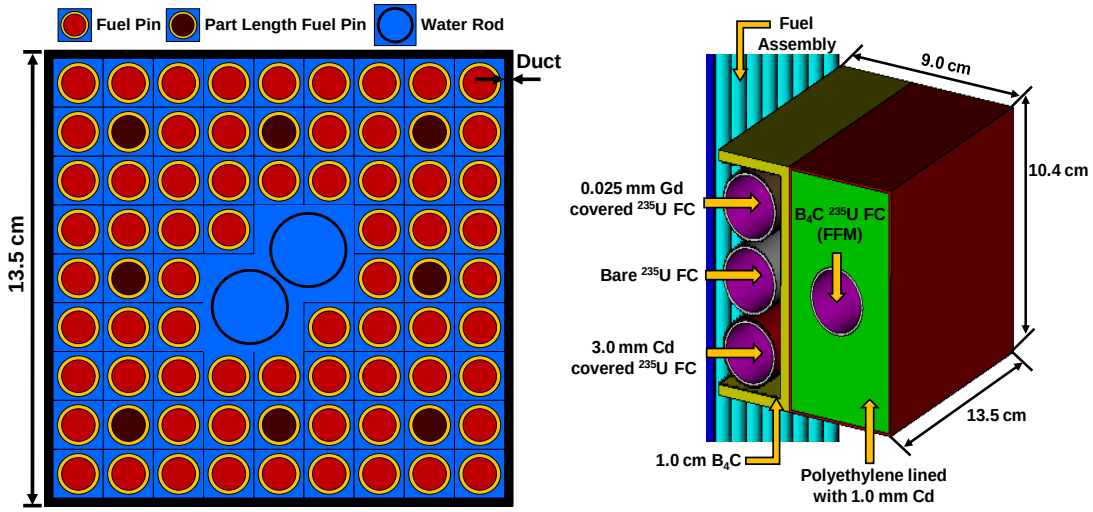
Assembly Data	
Lattice geometry	9 x 9 (square)
Assembly width (outer)	13.5 cm
Duct Thickness	0.25 cm
Fuel pin pitch	1.44 cm
Number of fuel pins	74 (8 Part-Length)
Inter-Assembly Gap	1.49 cm
Moderator	Light Water
Fuel Pin Data	
Fuel material	UO_2
Cladding material	Zircaloy-2
Initial ^{235}U Enrichment	3% and 4.5% ^{235}U
Fuel pellet density	10.01 g/cm^3
Fuel pellet diameter	0.975 cm
Outer pin diameter	1.118 cm
Cladding Thickness	0.071 cm
Active Fuel Height	371 cm
Partial Pin Fuel Height	244 cm

Table 1: Specifications for BWR 9x9 spent fuel assembly.

In contrast to a PWR, the light water coolant in a BWR is allowed to boil as it is circulated from the bottom to the top of the reactor. This results in the formation of vapor bubbles or voids in the upper region of a BWR. Voids displace part of the coolant and lower its density which in turn reduces the reactivity of the core. Since voids form in the upper region of a BWR, the moderating power is highest in the bottom region of the core (0% VF) [13]. Accounting for the different void fractions in a BWR is important to SINRD because it significantly affects the fuel burnup and thus the spent fuel isotopics we are trying to measure.

A top-down view of the BWR 9x9 fuel assembly (a) and the SINRD detector configuration (b) modeled in MCNPX are shown in Figure 2. SINRD consists of four FCs: Bare ^{235}U FC, boron carbide (B_4C) ^{235}U FC (located behind B_4C shield), 0.025-mm Gd covered ^{235}U FC, and 3.0-mm Cd covered ^{235}U FC. It should be noted that throughout the rest of this paper, we refer to the B_4C ^{235}U FC as FFM (or Fast Flux Monitor). The SINRD detector unit is located adjacent to the fuel assembly and approximately 10.4-cm high, 9.0-cm long, and 13.5-cm wide. The FFM is located behind the B4C shield to eliminate the resonance energy neutrons emitted from the fuel assembly. To increase counting statistics, the FFM was embedded in polyethylene to thermalize the fast neutrons that penetrated the boron shielding. The polyethylene was covered with 1.0-mm of Cd to reduce the background from thermal neutrons reentering the SINRD unit. The neutron flux entering the detector pod was measured using two FCs. The Bare ^{235}U FC was used to monitor the entire neutron flux spectrum with thermal neutron domination, and the FFM was used to monitor the fast neutron flux above the B_4C absorption cutoff

energy (3.8-keV). A fissile loading of 1.5-mg/cm^2 was modeled in the SINRD FCs using a 2-layer deposit thickness typical of standard commercial fission chambers. The ^{235}U FCs contained 93 wt% ^{235}U metal (19.1-g/cm^3) and the ^{239}Pu FCs contained 94 wt% ^{239}Pu metal (19.8-g/cm^3) [8].



(a) BWR 9x9 Fuel Assembly **(b) SINRD Detector Configuration**

Figure 2. (a) Top-down view of BWR 9x9 fuel assembly, (b) SINRD detector configuration modeled in MCNPX.

Ratios of different fission chambers were used to reduce the sensitivity of the measurements to extraneous material present in fuel (e.g. fission products). This also reduces the number of unknowns we are trying measure because the neutron source strength and the detector-fuel assembly coupling cancels in the ratio. It is important to note that SINRD requires calibration with a reference assembly of similar geometry. However, since this densitometry method uses the ratios of different FCs, most of the systematic errors related to calibration and positioning cancel in the ratios. In addition, SINRD can be calibrated with a fresh fuel assembly because it is not sensitive to neutron absorbing fission products in spent fuel [8,10].

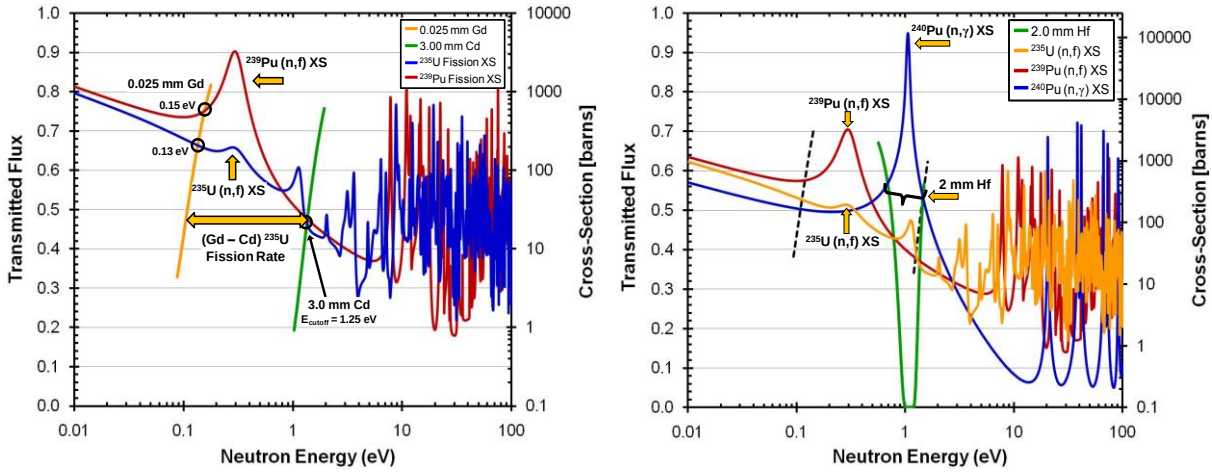
3. Analysis of SINRD for ^{239}Pu and ^{235}U Measurements

The SINRD detector configuration was optimized for quantifying ^{239}Pu and ^{235}U in a BWR 9x9 spent LEU fuel assembly with 0%, 40%, and 70% void fractions. To assess the sensitivity of SINRD to changes in the distribution of Pu isotopics in the spent fuel, we varied the initial enrichment (IE) from 3% to 4.5% ^{235}U. The cooling time was fixed at 5-yrs. The use of Gd and Cd ^{239}Pu FCs was investigated to determine how using ^{239}Pu FCs affects the sensitivity of the SINRD detector ratios to the ^{239}Pu content in spent fuel. This is important for LEU spent fuel because the ^{239}Pu and ^{235}U fractions are nearly equal at burnups greater than 30-GWd/MTU. However, it is important to note that ^{239}Pu FCs are not commercially available and would have to be specially manufactured. This could greatly increase the overall cost of SINRD. Therefore, we also investigated the use of all ^{235}U FCs in SINRD to quantify ^{239}Pu and ^{235}U in BWR spent LEU fuel.

3.1. SINRD Results for Quantifying ^{239}Pu

Different thicknesses of absorber filters were used in the SINRD ratios to determine the optimum ratio that maximized the SINRD signature for quantifying ^{239}Pu in BWR spent LEU fuel. Figure 3(a) shows how the large ^{239}Pu resonance at 0.3 eV can be windowed in energy by using the (Gd – Cd) ^{239}Pu fission rate based on the location of Gd and Cd absorption cut-off energies relative to the ^{239}Pu and ^{235}U fission cross-sections. The thick Cd filter (3-mm) absorbs the majority of neutrons in the low energy region of the ^{239}Pu resonance whereas the thin Gd filter (0.025-mm) transmits the majority of these lower energy neutrons. To determine how the absorption of low energy neutrons by ^{240}Pu affects

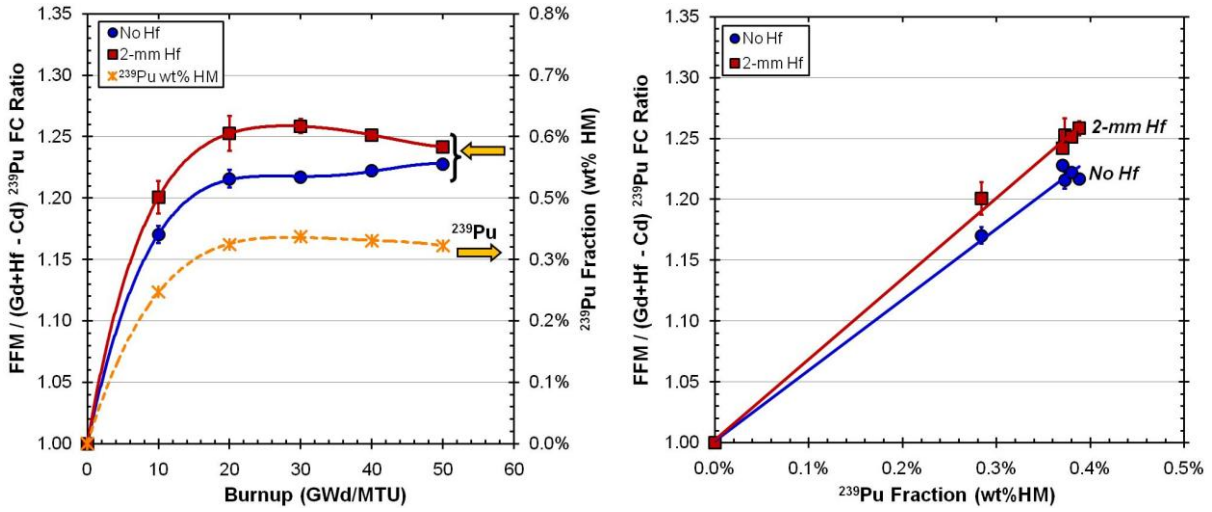
the SINRD FC ratio, a 2-mm Hf filter was added inside the Gd filter. The transmitted flux through a 2-mm Hf filter relative to the ^{240}Pu (n,γ) cross-section is shown in Figure 3(b).



(a) (Gd – Cd) energy window for ^{235}U & ^{239}Pu **(b) Transmitted flux through 2.0 mm Hf**

Figure 3. (a) ^{235}U and ^{239}Pu fission cross-sections within the (Gd – Cd) absorption cut-off energy window and (b) transmitted flux through 2-mm Hf relative to ^{240}Pu (n,γ) cross-section.

In Figure 4, the effect of using 2-mm Hf on FFM / (Gd – Cd) ^{239}Pu FC ratio is shown as a function of (a) burnup and (b) ^{239}Pu fraction. These results have been normalized to the fresh fuel case (3% IE). Using the FFM / (Gd – Cd) ^{239}Pu FC ratio, increased the slope of the SINRD signature by 18% compared to using just the FFM / Gd ^{239}Pu FC ratio. Adding 2-mm Hf to the Gd ^{239}Pu FC further increased the slope of the SINRD signature by 6%. Thus, the FFM / (Gd+Hf – Cd) ^{239}Pu FC ratio improved the SINRD signature by 24% overall. This is because the Hf filter absorbs the majority of neutrons in the same energy region as the ^{240}Pu (n,γ) resonance reducing the ^{240}Pu effect on the SINRD ratio within the (Gd – Cd) energy window.



(a) (Gd+Hf – Cd) ^{239}Pu Ratio vs Burnup **(b) (Gd+Hf – Cd) ^{239}Pu Ratio vs ^{239}Pu**

Figure 4. Optimized SINRD ratio for ^{239}Pu: FFM / (Gd+Hf – Cd) ^{239}Pu FC ratio versus (a) burnup and (b) ^{239}Pu wt%HM with no Hf and 2-mm Hf.

Referring to Figure 4(a), it is important to note that the results for the SINRD ratio with 2-mm Hf closely follow the curve for the ^{239}Pu fraction in LEU spent fuel over the burnup range of 0 – 50-GWd/MTU. However, when no Hf is used the SINRD ratio continues to increase with burnup even though the

^{239}Pu fraction decreases for burnups >30-GWd. The purpose for plotting the (Gd+Hf – Cd) ^{239}Pu FC ratio results versus burnup in Figure 4(a) and ^{239}Pu fraction in (b) was to illustrate that similarity of the curves in (a) translates to linear curves in (b) when the SINRD ratio was plotted versus ^{239}Pu fraction. It should also be noted that these results have been normalized to the fresh fuel case because in practice SINRD could be calibrated using a fresh fuel assembly.

In BWR spent LEU fuel, the relative concentrations of ^{235}U, ^{239}Pu, and ^{240}Pu change significantly for different void fractions. Figure 5 shows the buildup of Pu isotopics in LEU spent fuel with (a) 0% VF and (b) 70% VF. It should be noted that in spent LEU fuel with 40% VF the buildup of ^{239}Pu continues to increase over the burnup range of 0 to 50-GWd/MTU where the ^{239}Pu fraction is equal to 0.59 wt%HM at 50-GWd. The concentration of ^{239}Pu in 40-GWd spent LEU fuel with no void [Figure 5(a)] increased by 35% for 40% VF and by 68% for 70% VF. Therefore, we examined the effect of different void fractions and initial enrichments on the optimized SINRD ratio for quantifying ^{239}Pu using ^{239}Pu FCs with 2-mm Hf and all ^{235}U FCs with no Hf.

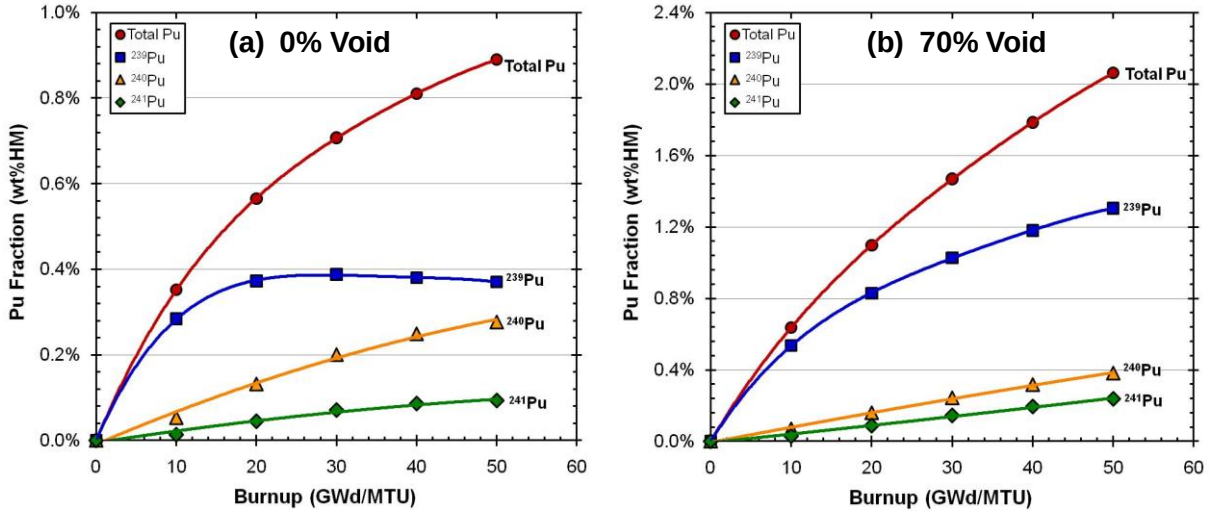


Figure 5. Buildup of Pu isotopics versus burnup in BWR spent LEU fuel with (a) 0% and (b) 70% void fractions (3% ^{235}U IE, 5-yrs cooled).

In Figure 6(a) and (b), the FFM / (Gd+Hf – Cd) ^{239}Pu FC ratio is compared to the FFM / (Gd – Cd) ^{235}U FC ratio versus ^{239}Pu fraction, respectively. These results were not normalized to the fresh fuel case. The maximum change in the SINRD ratio from varying the IE was 7.5% for the case with no void and all ^{235}U FCs; however, the sensitivity to IE decreases as the void fraction increases. Referring to Figure 6(b) for all ^{235}U FCs, the large scatter in the results for 0% and 40% void fractions indicates that ^{239}Pu FCs are needed to accurately measure the ^{239}Pu content in BWR spent LEU fuel especially for the case with 0% VF. This negative effect on our SINRD signature may be attributed to the fact that the concentration of ^{235}U relative to ^{239}Pu is large at low fuel burnups (≤ 30-GWd) and nearly equal at high burnups. As a result, the competing effects from the burnup of ^{235}U and buildup ^{239}Pu are wiping out our signature. Thus, all ^{235}U FCs cannot be used to determine the ^{239}Pu content in BWR spent LEU fuel with no void fraction. However, the results for 70% VF clearly show that all ^{235}U FCs can be used to quantify ^{239}Pu in spent LEU fuel. The ability to use all ^{235}U FCs to quantify ^{239}Pu in spent fuel with higher void fractions may be attributed to the fact that the amount of ^{239}Pu relative ^{235}U is much higher and that the buildup of ^{239}Pu continues to increase over burnup range of 0 to 50-GWd/MTU.

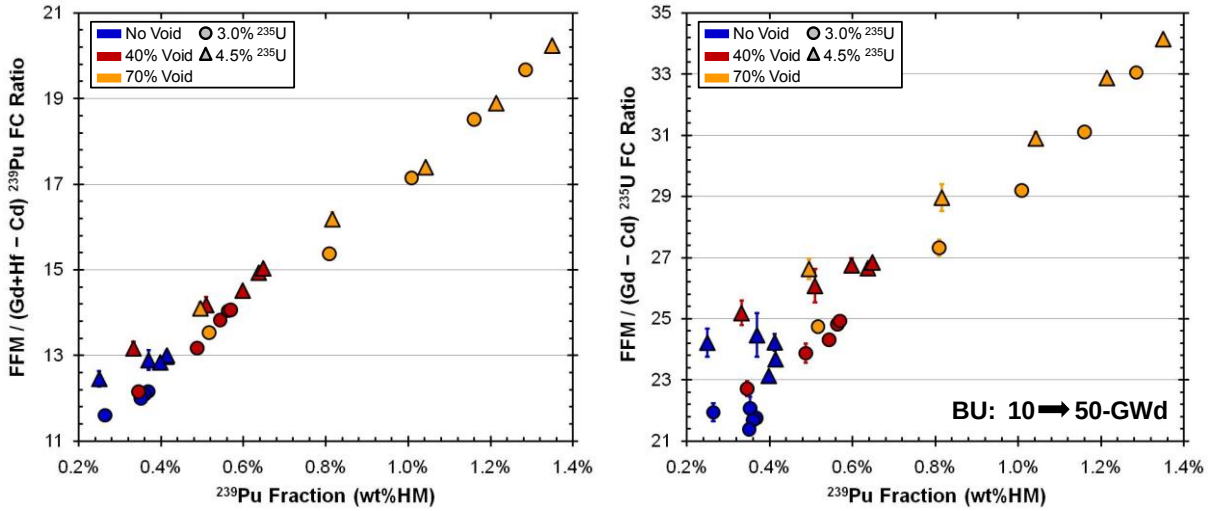


Figure 6. Comparison of (a) FFM / (Gd+Hf – Cd) ^{239}Pu FC ratio to (b) FFM / (Gd – Cd) ^{235}U FC ratio versus ^{239}Pu fraction for BWR spent LEU fuel with 3% and 4.5% ^{235}U IE.

It is important to note that the error bars shown on all results represent the calculated uncertainties in the SINRD ratios obtained via error propagations of expected counting statistics [8,10]. The expected count rates in the SINRD FCs are given in Table 2 for a BWR spent LEU fuel assembly with 40-GWd burnup (3% IE, 5-yrs cooled). The neutron source terms were 9.3E+07 n/s for 0% VF, 1.4E+08 n/s for 40% VF, and 1.9E+08 n/s for 70% VF. The use of Hf in the Gd ^{239}Pu and ^{235}U FCs reduced the count rate by 44% and 24%, respectively. The effect of using Gd and Cd ^{235}U FCs compared to ^{239}Pu FCs decreased the count rates in the Gd FC by 59% (no Hf) and Cd FC by 10%. Using error propagations, the lower count rates in the Gd and Cd ^{235}U FCs increased the relative uncertainty in the FFM / (Gd – Cd) ^{235}U FC ratio by 67% compared to using ^{239}Pu FCs. It is important to note that this increase in the relative uncertainty is significant because using all ^{235}U FCs also decreased the slope of the SINRD signature. It should also be noted that the expected count rates are somewhat conservative because the (α,n) contribution to the total neutron emission rate from the assembly was not accounted for. However, the (α,n) contribution is small compared to the spontaneous fission contribution to the total neutron emission rate especially at higher burnups.

SINRD Detectors	BWR Spent LEU Fuel [cps]		
	No Void	***40% Void***	***70% Void***
Bare ^{235}U	308 ± 0.29	415 ± 0.34	489 ± 0.37
FFM ^{235}U	896 ± 0.50	1451 ± 0.63	2280 ± 0.80
Gd ^{235}U	95 ± 0.16	145 ± 0.20	207 ± 0.24
Gd+Hf ^{235}U	72 ± 0.14	110 ± 0.17	158 ± 0.21
Cd ^{235}U	54 ± 0.12	86 ± 0.15	134 ± 0.19
Gd ^{239}Pu	241 ± 0.26	358 ± 0.32	479 ± 0.36
Gd+Hf ^{239}Pu	134 ± 0.19	199 ± 0.24	271 ± 0.27
Cd ^{239}Pu	60 ± 0.13	96 ± 0.16	148 ± 0.20

Table 2: Expected count rates in SINRD FCs for 40-GWd BWR spent LEU fuel for a fissile loading of 1.5-mg/cm^2 in the ^{235}U and ^{239}Pu FCs.

3.2. SINRD Results for Quantifying ^{235}U

The use of SINRD to quantify ^{235}U in LEU spent fuel was also investigated. The ability to measure ^{235}U using SINRD is important to verifying the burnup and initial enrichment of a BWR spent LEU fuel assembly. It is important to note that only ^{235}U FCs were used in SINRD for this analysis. Different

SINRD ratios were analyzed as a function of ^{235}U fraction to determine the best ratio for quantifying ^{235}U. The (Gd – Cd) ^{235}U / Bare ^{235}U FC ratio was the only ratio that linearly tracked ^{235}U in LEU spent fuel over burnup range of 0 to 50-GWd/MTU. Since ^{239}Pu also has a large fission resonance within the (Gd – Cd) energy window, it was also necessary to examine how resonance absorption by ^{239}Pu affects the (Gd – Cd) ^{235}U / Bare ^{235}U FC ratio. We determined that ^{239}Pu resonance absorption within the (Gd – Cd) energy window is contributing to our SINRD signature, especially at high void fractions, and thus should be accounted for. This was expected because the concentration of ^{239}Pu in BWR spent LEU fuel increases by a factor of 3 from 0% to 70% VF.

Figure 7 shows the effect of different void fractions and initial enrichments on the (Gd – Cd) ^{235}U / Bare ^{235}U FC ratio versus the ^{235}U + ^{239}Pu fraction in BWR spent LEU fuel. These results were not normalized to the fresh fuel case. Varying the initial ^{235}U IE, changed the SINRD ratio by less than 5% over burnup range of 0 to 50-GWd/MTU and thus, is not sensitive to this parameter. For both 3% and 4.5% ^{235}U IE, the SINRD ratio linearly tracks the ^{235}U + ^{239}Pu content in LEU spent fuel with 0%, 40%, and 70% VF. It should be noted that the slope of the SINRD FC ratio signature for quantifying ^{235}U + ^{239}Pu using all ^{235}U FCs decreased by a factor of ~9 for 0% VF, ~13 for 40% VF and ~16 for 70% VF compared to the slope of the SINRD ratio for quantifying ^{239}Pu using ^{239}Pu FCs. This effect is attributed to the fact that the ^{239}Pu fission cross-section is an order of magnitude larger than the ^{235}U cross-section within the (Gd – Cd) energy window. As a result, ^{239}Pu FCs have a higher sensitivity to ^{239}Pu resonance absorption in spent fuel.

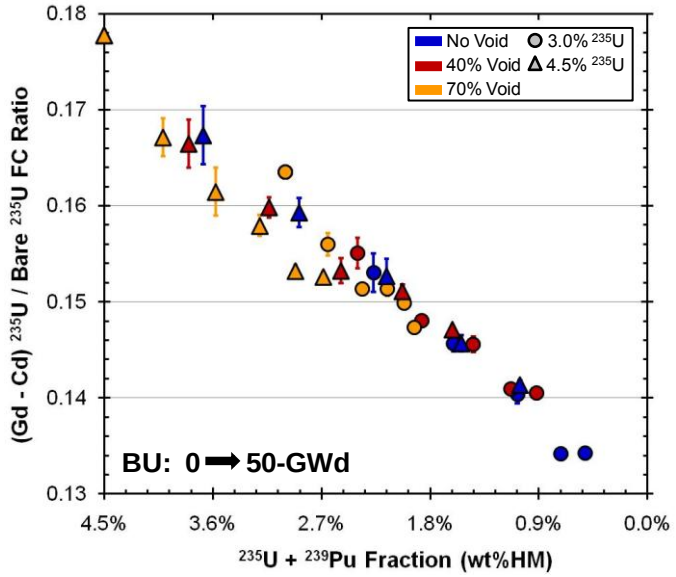


Figure 7. Comparison of (Gd – Cd) ^{235}U / Bare ^{235}U FC ratio versus ^{235}U + ^{239}Pu fraction in BWR spent LEU fuel.

4. Analysis of SINRD for Possible Diversion Scenarios

The sensitivity of SINRD to possible diversion scenarios was assessed for a BWR 9x9 spent LEU fuel assembly with 0%, 40%, and 70% void fractions. It is important to note that only ^{235}U FCs were used in SINRD. We used the following safeguards detection criteria to evaluate SINRD for this analysis:

- Independent of the Operator's declaration of:
  - burnup, initial enrichment, cooling time, and void fraction
- Sensitive to fuel pin removal over entire burnup range.
- Able to distinguish fresh and 1-cycle MOX fuel from 3- and 4-cycle LEU fuel [see Ref. 8].
- Recognize that IAEA will likely need to use all ^{235}U fission chambers.

4.1. Verification of Burnup

In a BWR 9x9 spent LEU fuel assembly, the ^{244}Cm spontaneous fission neutron emission rate is approximately 9.3E+07 n/s for burnup of 40-GWd/MTU with no void fraction. This source term is

further amplified by a factor of 2 – 3 by neutron multiplication in the assembly when in water. For spent LEU fuel, this high neutron source term provides adequate counting statistics in the fission chambers to give better than 1% precision in a few minutes for the SINRD ratios.

The use of SINRD to verify the burnup of a BWR spent LEU fuel assembly was investigated. In Figure 8, the ^{235}U and ^{244}Cm fractions are compared to the (Gd – Cd) ^{235}U / Bare ^{235}U FC ratio and FFM fission rate versus burnup for the diversion scenario where the burnup is misdeclared low. These results were normalized to the fresh fuel case with 4.5% IE. Since the ^{239}Pu content increases with burnup up to 30-GWd in LEU spent fuel, a proliferator is more likely to misdeclare the burnup low. Comparison of the results in Figure 8 (a) to (b), clearly shows that the FFM fission rate is directly proportional to ^{244}Cm and that the (Gd – Cd) ^{235}U / Bare ^{235}U FC ratio is proportional to ^{235}U in LEU spent fuel over the burnup range of 0 to 50-GWd/MTU.

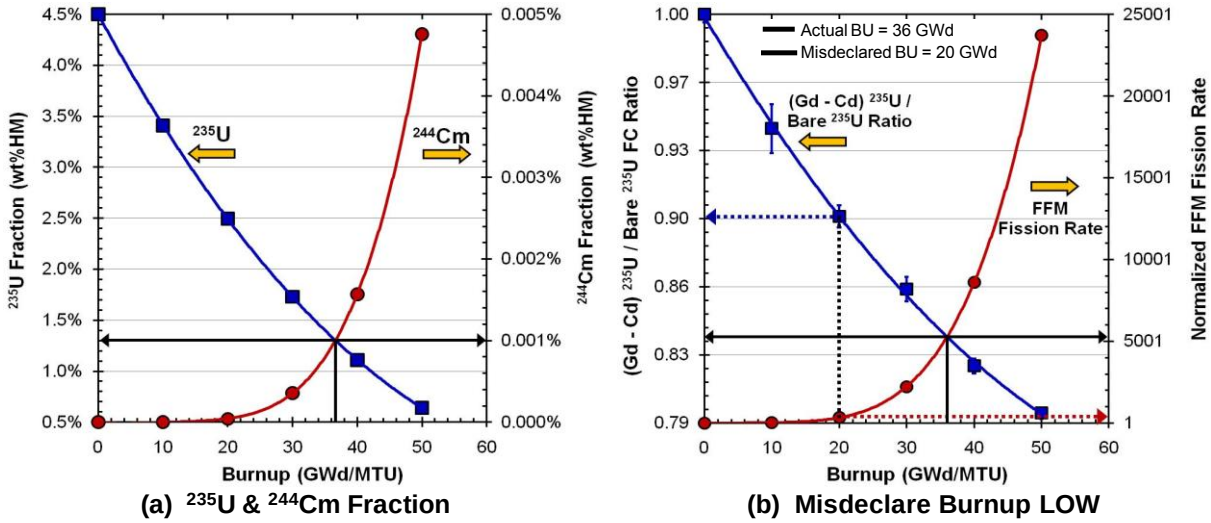


Figure 8. Comparison of (a) ^{235}U and ^{244}Cm fraction to (b) the (Gd – Cd) ^{235}U / Bare ^{235}U FC ratio and FFM fission rate versus burnup for diversion scenario where burnup is misdeclared low.

The fact that ^{235}U fraction decreases as a function of burnup, whereas the ^{244}Cm fraction increases enables us to verify the burnup of the BWR spent LEU assembly because the proliferator can only get one of these curves right by misdeclaring the burnup. Referring to Figure 8(b), the solid black line indicates the actual burnup of the assembly (36-GWd) and the solid black arrows point to the expected measured values at this burnup. The misdeclared burnup (20-GWd) is shown by the black dotted line. The dotted red and blue lines correspond to the expected measured values for the misdeclared burnup. When the burnup is misdeclared, the expected measured signals move in opposite directions. Thus, comparing a set of measurements where the burnup is misdeclared to a reference measurement with known burnup would clearly indicate an anomaly in the declaration.

4.2. Sensitivity of SINRD to Partial Defects

To assess the sensitivity and penetrability of SINRD, partial defects were modeled in a BWR 9x9 spent LEU fuel assembly with fuel burnups of 10 and 40-GWd/MTU. We uniformly removed 4 and 18 fuel pins (5% and 24% of the total pins, respectively) from two different radial regions of the assembly and replaced them with DU pins. The initial fuel enrichment was fixed at 3% ^{235}U for this analysis. The fuel pin removal locations of partial defects for Regions 1 and 2 are shown in Figure 9. Region 1 consists of the second row from the outer surface of assembly and Region 2 consists of rows in the center of the assembly. The average depth from the outer surface is 2.16-cm for Region 1 and 5.75-cm for Region 2.

To assess the penetrability of SINRD to partial defects, the percent change in the SINRD ratios was calculated for each region to determine if the diverted pins can be detected within 3σ uncertainty. The count times used for the diversion cases are given in Table 3. These count times are somewhat

conservative because they do not account for the contribution to the total neutron emission rate from (α,n) neutrons.

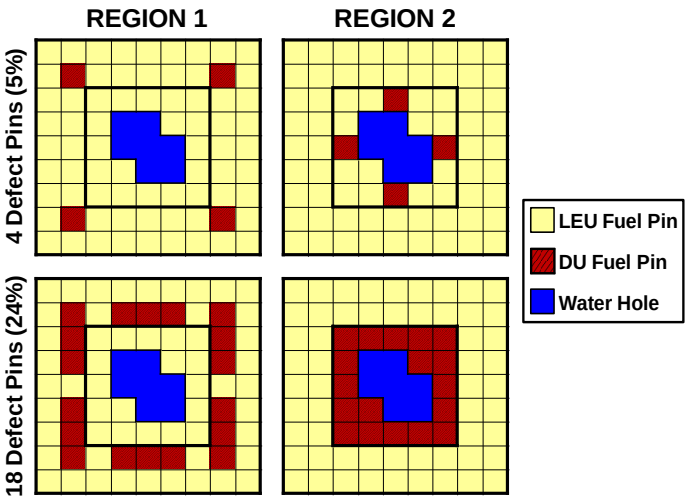


Figure 9. Fuel pin removal locations of defects for Regions 1 and 2 in BWR 9x9 assembly where red pin locations represent fuel pins that were removed and blue locations are water holes.

Void Fraction	Burnup [GWd/MTU]	
	10-GWd	***40-GWd***
No Void	5 hours	20 minutes
40% Void	4.5 hours	10 minutes
70% Void	4.5 hours	10 minutes

Table 3: Count times used to detect pin diversions within *3σ* uncertainty for BWR spent fuel.

The sensitivity of different SINRD ratios with 5% and 24% of the total number of pins removed from Regions 1 and 2 is given in Table 4 for BWR spent LEU fuel. The highlighted values correspond to the maximum positive and negative percent change in ratios that are within *3σ* uncertainty for 5% and 24% pins removed from each region. The cells that are shaded gray correspond to the percent in change the detector ratios that are not within *3σ* uncertainty of a spent fuel assembly with no diverted pins. It should be emphasized that all ^{235}U FCs and no Hf were used to obtain these results.

It is important to note that for a BWR spent LEU fuel assembly with burnup of 10-GWd/MTU, none of the SINRD ratios can detect the diversion of 4 pins (5%) from Region 2 within *3σ*. If the count time was increased to 40-hrs for 0% void, 12-hrs for 40% void, and 25-hrs for 70%, then only the FFM / Bare ^{235}U FC ratio could detect 5% pin diversions within *3σ* in Region 2. Error propagations [8,10] were used to calculate the uncertainties in the percent change in the SINRD ratios for all diversion cases. These uncertainties were between 0.2% – 1% for the FFM / Bare ^{235}U FC ratio using the count times given in Table 3. Thus, this type of measurement could show the departure from a reference fuel assembly with no defects. A summary of the results shown in Table 4 is given below:

- All SINRD ratios have the highest sensitivity to pin removal in Region 1.
- For BWR spent LEU fuel, the FFM / Bare ^{235}U FC ratio is the most sensitive SINRD ratio for detecting fuel pin diversions within *3σ* from Regions 1 and 2.
  - This ratio is sensitive to reactivity changes in the fuel assembly due to changes in the concentration of thermal absorbers.
  - The percent change in this ratio is positive for pin removal from Regions 1 and 2.

Region Defects	Burnup	SINRD Ratios *BWR Spent LEU*	REGION 1			REGION 2		
			0% Void	*40% Void*	*70% Void*	*0% Void*	*40% Void*	*70% Void*
5% Pin Defects (4 pins)	***10 GWd***	FFM / (Gd - Cd) ^{235}U	*2.13%*	*2.35%*	*3.21%*	*-0.59%*	*0.70%*	*-0.16%*
		FFM / Bare ^{235}U	**4.49%**	**4.85%**	**4.67%**	*1.07%*	1.53%	0.75%
		Bare ^{235}U / Gd ^{235}U	*-3.51%*	*-3.56%*	-3.21%	*-1.81%*	*-1.86%*	*-1.14%*
		Bare ^{235}U / Cd ^{235}U	*-4.32%*	*-4.25%*	**-4.38%**	*-1.91%*	*-2.62%*	*-1.30%*
	40 GWd	FFM / (Gd - Cd) ^{235}U	2.87%	**5.20%**	4.13%	*0.92%*	*1.71%*	*1.10%*
		FFM / Bare ^{235}U	**3.76%**	4.60%	**4.84%**	0.86%	1.24%	0.97%
		Bare ^{235}U / Gd ^{235}U	-2.38%	-2.50%	-3.01%	-1.13%	-1.35%	-1.35%
		Bare ^{235}U / Cd ^{235}U	**-3.54%**	**-4.73%**	**-4.29%**	**-2.07%**	**-2.62%**	**-2.17%**
24% Pin Defects (18 pins)	***10 GWd***	FFM / (Gd - Cd) ^{235}U	*9.03%*	*9.10%*	11.3%	*-1.60%*	*-1.28%*	*0.72%*
		FFM / Bare ^{235}U	**15.6%**	**14.3%**	**14.7%**	**4.58%**	**5.01%**	**5.33%**
		Bare ^{235}U / Gd ^{235}U	-13.8%	-11.8%	-11.5%	-8.84%	-9.09%	-8.77%
		Bare ^{235}U / Cd ^{235}U	**-18.8%**	**-16.4%**	**-17.1%**	**-10.7%**	**-11.0%**	**-11.6%**
	40 GWd	FFM / (Gd - Cd) ^{235}U	9.03%	11.5%	12.7%	*-0.30%*	*1.68%*	*0.83%*
		FFM / Bare ^{235}U	**12.9%**	**14.8%**	**16.3%**	**4.21%**	**5.67%**	**6.73%**
		Bare ^{235}U / Gd ^{235}U	-10.1%	-11.5%	-12.9%	-7.66%	-8.52%	-10.0%
		Bare ^{235}U / Cd ^{235}U	**-14.9%**	**-17.4%**	**-18.2%**	**-10.1%**	**-11.6%**	**-12.1%**

Table 4: Percent change in SINRD ratios with 5% and 24% fuel pins removed from Regions 1 and 2 for BWR spent LEU fuel (No Hf).

In Figure 10, the fuel pin removal results for FFM / Bare ^{235}U FC ratio as a function diversion case are shown for BWR spent LEU fuel with no void fraction and burnup of (a) 10-GWd and (b) 40-GWd. The solid line represents the signal from the case with no diversions; the dashed lines represent ±1% change in the SINRD ratio to account for systematic errors. We chose to use the FFM / Bare ^{235}U FC ratio in this analysis because it was the most sensitive ratio for detecting fuel pin diversions within 3σ from Regions 1 and 2. These results show that the SINRD ratio has the highest sensitivity to fuel pin diversions from Region 1. The diversion of 4 pins (5% of total number of pins) from Region 2 for both 10 and 40-GWd/MTU are the only cases that are not clearly within ±1% of the no diversion signal.

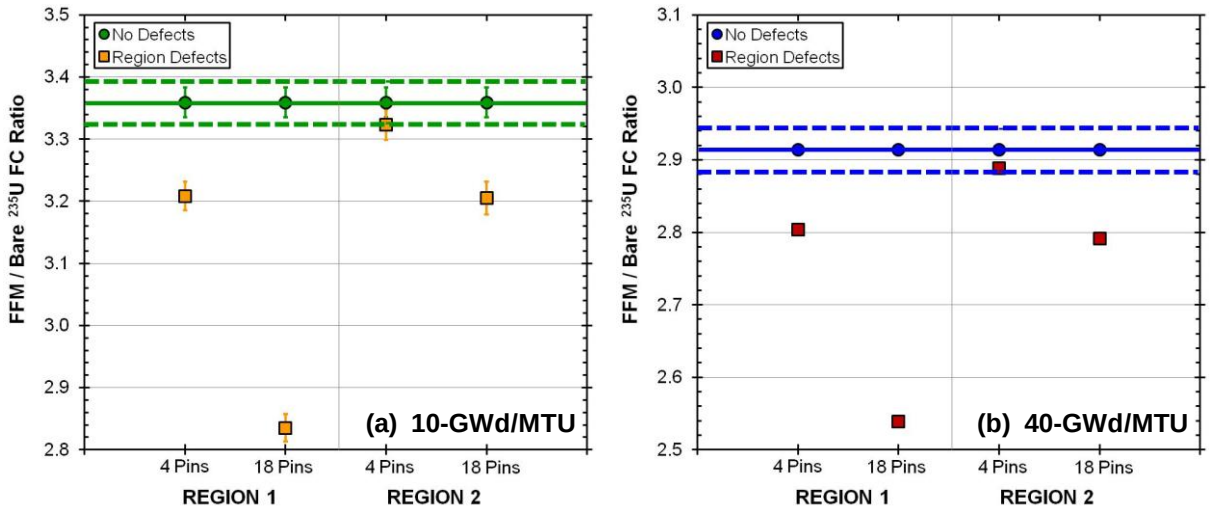


Figure 10. Pin removal results for FFM / Bare ^{235}U FC ratio as a function diversion case for BWR spent LEU fuel with burnup of (a) 10-GWd and (b) 40-GWd (no void fraction).

5. Conclusions

We have simulated the change in different SINRD detector ratios over a burnup range of 0 – 50 GWd using MCNPX. SINRD provides an excellent response for quantifying ^{239}Pu using ^{239}Pu FCs and total fissile content (^{235}U+^{239}Pu) using ^{235}U FCs in a BWR spent LEU fuel assembly with void fractions of 0%, 40%, and 70%. For determining ^{239}Pu, the FFM / (Gd+Hf – Cd) ^{239}Pu FC ratio was optimized using 2-mm Hf. This SINRD ratio is proportional to the ^{239}Pu mass in the assembly over the burnup range of 0 to 50-GWd. Due to the fact that the IAEA might require all ^{235}U FCs, the use of the FFM / (Gd – Cd) ^{235}U FC ratio to determine ^{239}Pu was also investigated. All ^{235}U FCs cannot be used to quantify the ^{239}Pu content in BWR spent LEU fuel with 0% void fraction but can be used for 40% and 70% void fractions. The ability to use all ^{235}U FCs to quantify ^{239}Pu in spent fuel with 40% and 70% void fractions may be attributed to the much larger amount of ^{239}Pu relative ^{235}U and that the ^{239}Pu content continues to increase over burnup range of 0 to 50-GWd/MTU. However, all ^{235}U FCs can be used to quantify the total fissile content in LEU spent fuel using the (Gd – Cd) ^{235}U / Bare ^{235}U FC ratio. This SINRD ratio is proportional to the (^{235}U + ^{239}Pu) mass in the assembly over the entire burnup range and is relatively insensitive to initial enrichment and void fraction.

The sensitivity and penetrability of SINRD was assessed by modeling partial defects in a BWR 9x9 spent LEU fuel assembly. It is important to note that all ^{235}U FCs were used in this analysis. The percent change in the SINRD ratios was calculated for Regions 1 and 2 to determine if the diverted pins can be detected within 3σ. For a BWR spent LEU fuel assembly with burnup of 10-GWd/MTU, none of the SINRD ratios can detect 5% pin diversions in Region 2 within 3σ. Thus, SINRD would need to be integrated with another verification technique to ensure no pins were diverted from the center of the assembly. Based on the results from these calculations, the FFM / Bare ^{235}U FC ratio is the best ratio for detecting pin diversions from a BWR spent LEU fuel assembly. This is because the FFM / Bare ^{235}U FC ratio has the lowest uncertainty of all the SINRD ratios which is important for spent LEU fuel because neutron source term is very low at low burnups (<20-GWd). These uncertainties were between 0.2% – 1% for the FFM / Bare ^{235}U FC ratio. Thus, this type of measurement could show the departure from a reference fuel assembly with no defects.

The purpose of the BWR spent fuel simulations was to assess the ability of SINRD to measure the fissile content in spent fuel and the sensitivity and penetrability of SINRD to partial defects in an assembly. Based on the results from these simulations, we have concluded that SINRD provides a number of improvements over current IAEA verification methods. These improvements include:

1) SINRD provides absolute measurements of burnup independent of the operator's declaration.
2) The sensitivity of SINRD to initial enrichment is small (<5%) and can therefore be used at multiple spent fuel storage facilities.
3) The calibration of SINRD at one reactor facility carries over to reactor sites in different countries because it uses the ratio of FCs that are not facility dependent.
4) SINRD is sensitive to pin removal over the entire burnup range and can verify the diversion of 5% of fuel pins within 3σ for BWR spent LEU fuel with burnups >25-GWd/MTU.

6. Acknowledgements

We would like to acknowledge the Department of Energy National Nuclear Security Administration's Office of Global Security Engagement and Cooperation (NA-242) and the Office of Nuclear Safeguards & Security (NA-241) for their support in the development of the SINRD method. This work was funded under the NA-24 Next Generation Safeguards Initiative (NGSI) program.

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X-Ray Fluorescence for Safeguards of Spent Fuel Assemblies

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Abstract:

Quantifying the plutonium (Pu) content in spent fuel is necessary for many reasons, particularly to verify that diversion or other illicit activities have not occurred. The Next Generation Safeguards Initiative (NGSI) is funding a large collaborative effort between multiple laboratories and universities to improve spent nuclear fuel safeguards methods and equipment. This effort involves the current work of modeling several different nondestructive assay (NDA) techniques. We expect that the best solution, given the safeguard need, will involve combining a few instruments into a system. The insights gained from this research will be used to down-select a few of the most promising techniques that complement each other from the original set. The goal is to integrate the selected instruments together to create an accurate measurement system for fuel verification that is also robust enough to detect diversions. These instruments will then be, fabricated and tested. This work examines one of the NDA techniques—measuring x-ray emission peaks from Pu and uranium (U) to gather information about their relative quantities in a spent fuel assembly. X-ray fluorescence (XRF) is unique among the techniques in that it is the only one able to give the ratio of elemental Pu/U, allowing the Pu mass to be quantified for the assembly if U is known. XRF also presents many challenges, such as penetration issues because the low energy x-rays desired are effectively shielded by the first few millimeters of a fuel pin. This paper explores the capability of the Monte Carlo N-Particle eXtended (MCNPX) transport code to predict XRF measurements of spent fuel rods made at Oak Ridge National Laboratory (ORNL). MCNPX is then used to simulate detected x-ray peaks for a suite of modeled spent fuel assemblies with various burnups (BUs), initial ^{235}U enrichments (IEs), and cooling times (CTs). Analysis of XRF feasibility for safeguards of spent nuclear fuel assemblies will be discussed.

Keywords: X-Ray; Fluorescence; Spent Fuel; NDA;

1. Introduction

The U.S. Department of Energy (DOE) through the Next Generation Safeguards Initiative (NGSI) of NA-24 is funding a large collaborative effR. ort between multiple laboratories and universities to improve spent nuclear fuel safeguards methods and equipment [1].This effort involves the current work of modeling several different nondestructive assay (NDA) techniques. Several approaches are being researched since we expect that the safeguard need will best be met by combining the attributes from a few NDA techniques [2]. The understanding gained from this effort will be used to down-select to a few of the most promising techniques that complement each other most effectively. The goal is to integrate the selected instruments together to create an accurate measurement system for fuel verification that can also detect diversions. These instruments will be fabricated and tested to directly demonstrate fitness for purpose.

The work presented here focuses on the feasibility of using x-ray fluorescence (XRF) as a tool to quantify the elemental abundance of plutonium (Pu) in pressurized water reactor (PWR) spent fuel assemblies. XRF is unique among the other techniques being explored in that it offers the ability to measure the elemental quantity of Pu, as opposed to isotopic properties. Given that our motivation is to apply XRF to assemblies, recent XRF measurements of single spent fuel pins provide an opportunity to build confidence that XRF modeling capabilities can accurately predict the x-ray signal from spent fuel. Therefore, the first task will be to attempt to simulate pin measurements, compare to measured data, and then simulations will examine the feasibility of the XRF technique on assemblies.

1.1 Background

When an electron is freed from the inner orbit of an atom, an electron from a less tightly bound orbit transitions to the inner orbit to fill the vacancy. The difference in binding energy between the two orbits is released as an x-ray with high probability. The energy of the x-rays is characteristic of the element. Therefore, measuring the ratio of uranium (U) and Pu x-ray peaks provides information about the relative concentration of U and Pu present in the sample. If the total mass of U is accurately known, then the total mass of Pu can be calculated. There are many means by which an atom can become ionized in spent fuel. The two main sources of excitation energy, those considered in this work, are gamma rays and beta particles, both of which have a high passive flux inside the irradiated fuel arising from the decay of fission fragments. The energies of x-rays for Pu and U for the most intense peaks (K_α and K_β) are shown in Table 1 [3]. The most intense $K_{\alpha 1}$ x-rays are used for the Pu / U ratio.

	Levels	Energy [keV]		Relative Intensity	
X Ray	(Final - Initial)	Uranium	Plutonium	Uranium	Plutonium
$K_{\alpha 2}$	K - L_2	94.67	99.55	61.9	62.5
$K_{\alpha 1}$	K - L_3	98.44	103.76	100	100
$K_{\beta 1}$	K - M_3	111.3	117.26	22	22.2
$K_{\beta 2}$	K - N_{2-5}	114.5	120.6	12.3	12.5
$K_{\beta 3}$	K - M_2	110.41	116.27	11.6	11.7

Table 1: Energies and relative intensity of major x-rays for U and Pu.

1.2 Measuring XRF

Photon measurements of spent fuel using high resolution spectrometers exhibit a large background continuum in the low energy x-ray region due to Compton scattering of the intense energetic gamma photons created by spent fuel. The high Compton continuum can make x-ray measurements of the less abundant Pu difficult because of the relatively small signal-to-background ratio produced. In typical PWRs, only 1 to 2% of the spent fuel is Pu by weight. In lower burnup (BU) fuels with less Pu, the signal-to-background of the Pu x-ray peak may be too small for accurate and timely measurements. Few attempts have been made in the past to measure the Pu x-rays in spent nuclear fuel, [4] but recent measurement campaigns demonstrate Pu x-rays for PWR spent fuel pins with BUs ranging from 35 to 70 Gwd/tU can be measured [5]. One of the challenges XRF faces is a mean free path in nuclear fuel of ~0.5 mm. Therefore, only the outer layer of a fuel pin is really being measured. Furthermore, the high photon flux created by spent fuel means that long collimators are needed to keep count rates within detector limitations. In addition, because of the low signal-to-background of the Pu x ray, long count times are needed per detector; hence, it is necessary to use multiple detectors in order to achieve reasonable count times.

2. North Anna Fuel Pin Measurements

XRF experimental measurement campaigns of spent fuel were undertaken at Oak Ridge National Laboratory (ORNL) in May 2008, July 2008, and January 2009. Fuel pins from North Anna and Three Mile

Island were measured at various axial positions along the fuel rod to get a range of BUs. Data were collected using both coaxial and planar high purity germanium (HPGe) detectors to cover a wide range of energies with high resolution. The coaxial detector would be typical of the kind used for BU determination, but the thinner planar detector offers better signal-to-background for x-ray energies due to the low efficiency for higher energy gammas.

This section focuses on measurements of a North Anna fuel pin nominally burned to ~67 Gwd/tU, 4.7 years cooling time (CT), with 4.2% initial enrichment, which is used for comparison to Monte Carlo N-Particle eXtended (MCNPX) computational models [6]. While measurements are currently only available for single fuel pins, the ultimate goal of this project is to determine the capabilities of XRF to efficiently measure entire assemblies. As the first step to that end, modeling efforts are benchmarked against the ORNL fuel pin measurements. The North Anna fuel pin measurements were selected because of the high BU, and thus high Pu content; therefore, the benchmarked case represents the best case scenario.

2.1 XRF Experiment

The North Anna fuel pin, labeled 649, was cut axially into smaller pieces. The pin piece, labeled 649C, was cut far enough away from the ends that the axial Pu profile would be flat. 649C was wrapped in a stainless steel shipping tube with walls 1/16th of an inch thick (0.15875 cm). The pin piece was positioned inside a hot cell, and a planar HPGe detector was placed outside the hot cell at the end of a stainless steel collimator that penetrates through a concrete wall. The detector was ~120 cm from the center of the fuel pin. The collimator was ~90 cm long, with a 0.2 cm diameter hole. The collimator hole is not concentric with the collimator, which is 2.4 cm thick at the narrowest point. Figure 1 illustrates the experimental setup. Multiple measurements were taken with the planar detector using a count time of 1 hour, in which Pu x-ray peaks were visible. An overnight spectrum was also taken to improve statistical precision. Figure 2 shows the overnight spectrum of the North Anna fuel pin 649C taken at ORNL. The U x-ray peaks are clearly visible above the Compton background. A challenge of XRF for spent fuel is the relatively small number of counts in the Pu $K_{\alpha 1}$ peak. Fortunately, the continuum in the background region is fairly level.

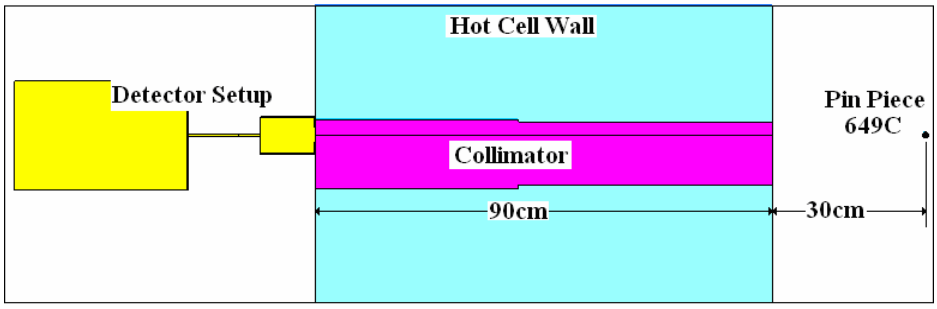

Figure 1: Schematic of experimental setup.

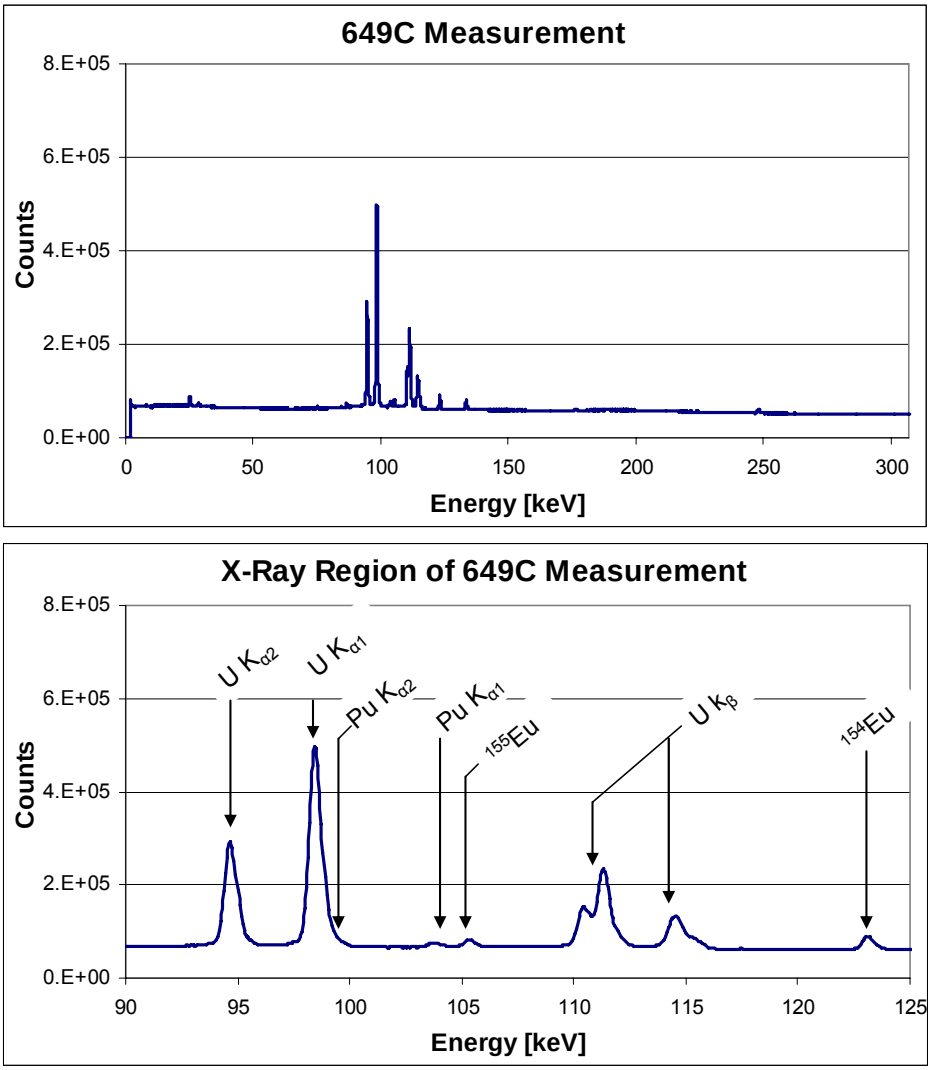

Figure 2: Measurement of pin 649C at ORNL with a planar HPGe detector. The live count was 44,263 seconds with 18% dead-time. Top: Full spectrum. Bottom: XRF region with peaks marked.

3.0 Simulating XRF Measurements of Fuel Pin 649C

The XRF measurements of North Anna spent fuel taken at ORNL presented an excellent benchmarking opportunity for the XRF models. MONTEBURNS/ORIGEN2 was used to calculate the isotopic inventories present in the fuel pin. The North Anna fuel pin had an initial enrichment of 4.2% and was burned to 67 Gwd/tU in three cycles with a final cool down of 4.7 years. The pellet radius was 0.40957 cm with a 0.00826 cm thick air gap and 0.05715 cm of cladding. Ten radial regions of decreasing thickness were used to capture the rapid increase of Pu concentration at the pin edge. Figure 3 shows the radial profile of Pu calculated for 649C.

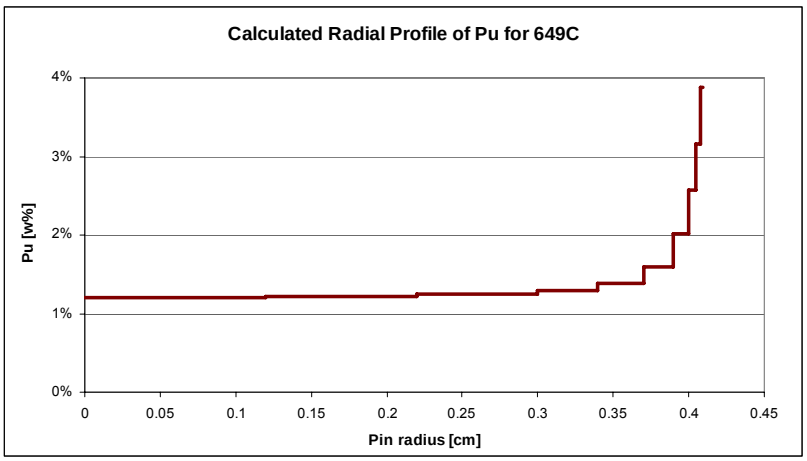

Figure 3: Radial profile of Pu calculated for pin 649C using MONTEBURNS/ORIGEN2.

3.1 Fluorescing Source Term

Only x-rays produced from beta and gamma excitation of the atom are considered in this work. The contribution of excitation from alpha particles and all other sources were considered negligible. Given that MCNPX modeling of electron transport for the determination of the XRF contribution from beta particles requires significant computational resources, research was done to see if simplifying assumptions could be made to minimize the amount of electron transport done. Therefore, simulations were done to determine the relative importance of the photon source and beta source in spent fuel as well as electron transport initiated by both of the aforementioned radiation sources.

The gamma source was calculated using a methodology and code that was developed for calculating delayed gamma source terms [7][8]. The code uses data libraries from CINDER'90 [9] for fission yields and reaction rates and uses Evaluated Nuclear Data Files (ENDF) for decay constants and branching ratios to produce a robust gamma source term from given isotopic inventories. Figure 4 shows the gamma source term created. Since the relative intensity of gammas spans many orders of magnitude, most of the gamma lines will be created too infrequently to be of consequence in the MCNPX calculation. The discrete gamma lines that failed to meet a relative probability of emission criteria were discarded in the source deck. The criteria was chosen based on the number of source particles sampled in MCNPX so that each photon source line that was kept was likely to be sampled at least a few hundred times throughout the run.

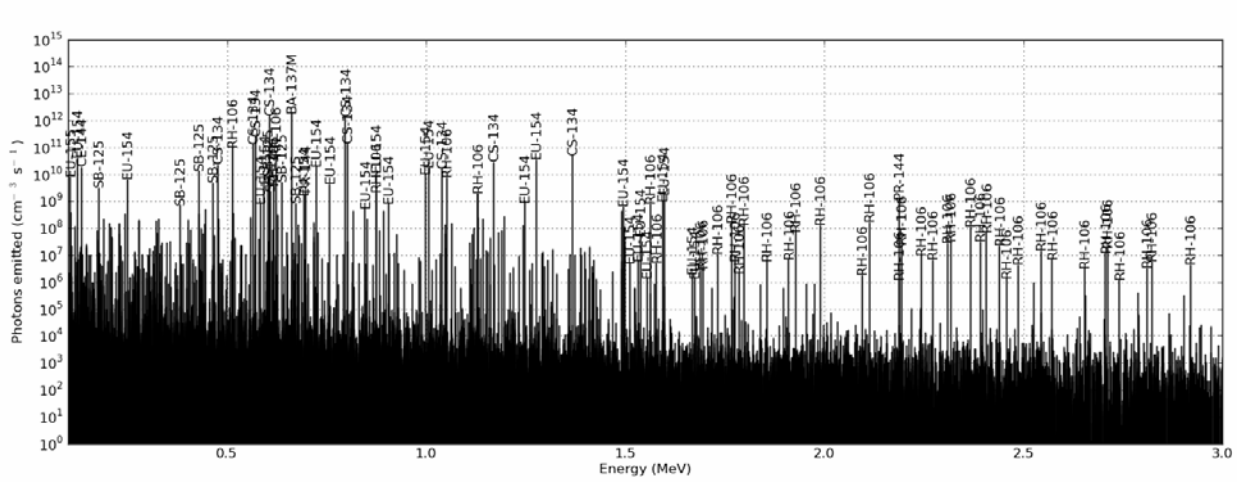

Figure 4: Gamma source term generated for gamma and beta comparison.

A literature search showed that comprehensive lists of beta emitters in spent fuel important to XRF are not well known. As a consequence, it is understood that while we think our beta source term has included the most important beta emitters that were tracked by the MONTEBURNS/ORIGEN2 code, it should be noted that this source term may be deficient. The intensity of beta particles emitted by each isotope in 1keV energy bins is available tabulated from Laurence Berkeley National Laboratory [10]. The combined binned energy spectrum for a single pin region is shown in Figure 5.

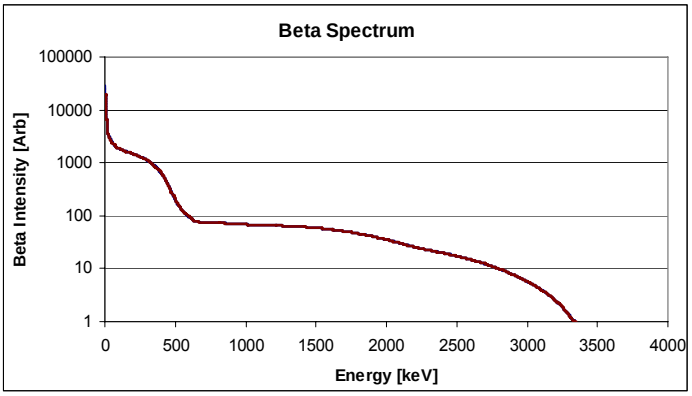

Figure 5: Calculated beta energy spectrum for a radial region in 649C.

A comparison of the x-ray signal strength induced by beta and gamma sources was performed to better understand the relative importance of both sources. For comparison, an MCNPX simulation was run of a 6 cm tall piece of fuel pin 649C. The photon signal exiting radially out of the stainless steel shipping tube was tallied in MCNPX for both source terms. Figure 6 shows the comparison between beta and gamma source terms in the XRF region. Calculations show that the majority of the contribution to the XRF signal is from the gamma source term. The beta source term was found to contribute roughly 10% to the XRF peaks. Since MCNPX transportation of beta induced XRF is orders of magnitude more computationally demanding than calculating its gamma counterpart, the remaining calculations will only involve the gamma term, with the understanding that a portion of the XRF signal is unaccounted for. The scenario simulated used the isotopic inventories calculated for fuel pin 649C. For our feasibility study, we have concluded that only simulating the gamma term will be sufficient to determine our key goal of relative Pu/U x-ray emission.

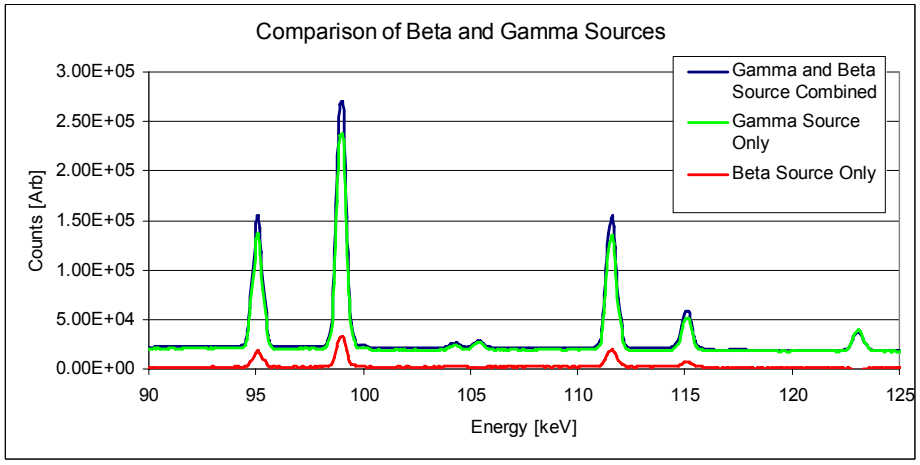

Figure 6: Comparison of beta induced and gamma induced XRF.

3.2 Radial Behavior of Source Term

An examination of what part of the fuel pin produces the measured U and Pu x-rays was conducted. Considering each of the ten regions modeled separately, the contribution to the U K_{a1} peak on a per source particle bases is plotted for gamma sources in Figure 7. Particles created closer to the outer radius of the fuel have a greater chance of contributing to the XRF signal. This seems intuitive as XRF signal that is created away from the outer edge is more likely to be attenuated. As the gamma source moves closer to the edge, however, the contribution to XRF begins to rapidly drop off. We interpret this as the gamma source at this point becomes more likely to escape itself than to create an x-ray to begin with.

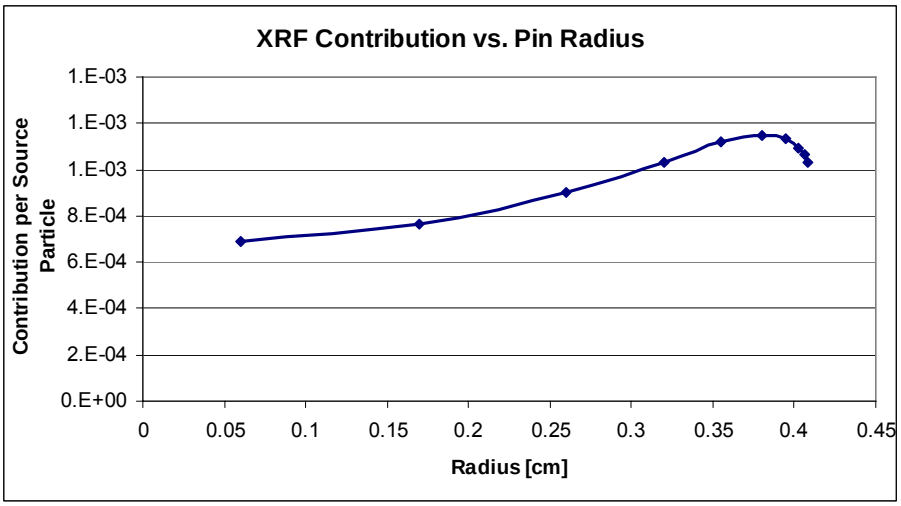

Figure 7: XRF signal that escapes the fuel pin by region for gamma source.

3.3 Modeling Methodology

To calculate the detector response in the HPGe crystal, the MCNPX f8 tally energy deposition is used. F8 tallies allow very limited variance reduction in MCNPX. Given that analog transport mode is required with f8 tallies, the number of particles that escape the fuel pin, travel down the long collimator, and interact in the detection crystal are too few and computationally inefficient for our resources. Therefore, we needed to make some simplifying assumptions. We decided to break the calculation into two pieces in order to bring the run-time down to a manageable range.

The first MCNPX model is a 6 cm tall fuel pin, including cladding and shipping tube, simulating 649C, to simulate the creation of x-rays inside the fuel. Only 6 cm of fuel pin is modeled because it was found that for a horizontal collimator with a radius of 0.1cm that is 5cm long centered on a 6 cm fuel pin, the probability of photons making it to the end of the collimator becomes negligible for sources in the fuel born about 2 cm above the center of the collimator. Therefore, modeling a longer fuel pin piece would not contribute to the MCNPX tally and would only require longer run times. The energy spectrum of photons exiting the shipping tube surface is tallied to approximate the energy distribution that is incident on the detector. Furthermore, collimator itself is modeled and the total number of photons that reach the end of the collimator is also tallied to approximate the fraction of photons that are incident on the detector per source gamma. The collimator is long enough that it is considered ideal, and thus, only the pin hole is modeled. This also significantly reduces the computational needs by not tracking multiple particle collisions and absorption in the collimator. Since the fuel pin is symmetric, the model is designed with multiple collimators like the spokes on a wheel to further increase the computational efficiency. The resulting total magnitude of every collimator is normalized by dividing by the number of collimators used. Figure 8 shows the MCNPX model with 180 collimators. Figure 9 shows a detailed look at the fuel pin modeled.

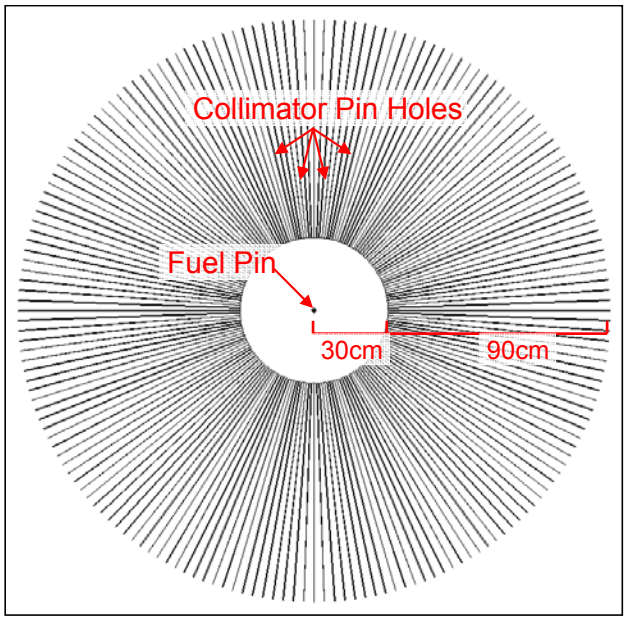

Figure 8: Diagram of MCNPX for the first part of the 649C experiment simulation.

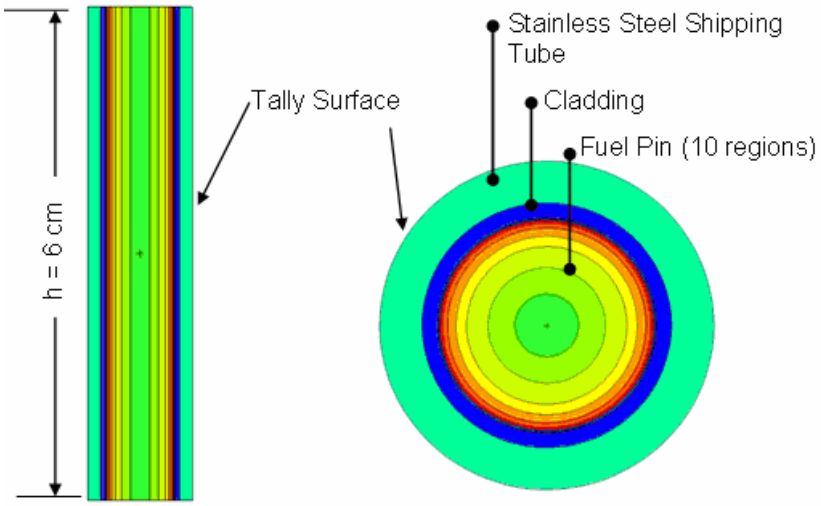

Figure 9: MCNPX geometry of the fuel pin.

Not all photons that exit the shipping tube were included in the MCNPX tally for energy spectrum. Photons created in the same location that exit the fuel pin with different angles to the surface normal cross different distances of material, and therefore are attenuated differently. Thus, tallying all photons that escape the fuel pin would unacceptably bias the tallied energy spectrum. The maximum angle that an ideal collimator, which is 90 cm long with a radius of 0.1 cm, allows is 0.127º. Tallying only photons that cross the surface normal near this angle is not computationally feasible. It was found that tallying only photons exiting the shipping tube within 20º of the surface normal offered sufficient tally statistics with a minimal bias to the energy spectrum. Therefore, the tallies used to approximate the energy spectrum exiting the shipping tube included photons with angles up to 20º to the surface normal.

The second part uses an MCNPX model focused on the detector response by tallying the spectrum of energy deposition in the HPGe crystal. The photon source particles are created perpendicular to the HPGe crystal with the energy distribution calculated in the first part. All uncertainties and angular information in the first simulation is lost. Since the detector face is much larger than the collimator pin

hole, the angular distribution of particles that make it to the end of the collimator is unnecessary. Furthermore, since the collimator is considered ideal, the Compton background from any photons that would have scattered off the collimator, but still contributed to detector counts are not included. The aluminum casing around the crystal, liquid nitrogen Dewar, cold finger, and the crystal structural casing inside the detector are included in the MCNPX model (Figure 10) for Compton background in the simulated detector. The HPGe crystal is 1.5 cm thick and 2.52 cm in diameter and is centered over the collimator hole. The MCNPX model includes a Gaussian Energy Broadening (GEB) card to account for the energy broadening properties of the HPGe detector. The final modeled spectrum is calculated by taking the simulated detector response from part two and normalizing it by the calculated total gamma source strength during the measurement live time multiplied by the fraction of source particles that reach the end of the collimator tallied in part 1.

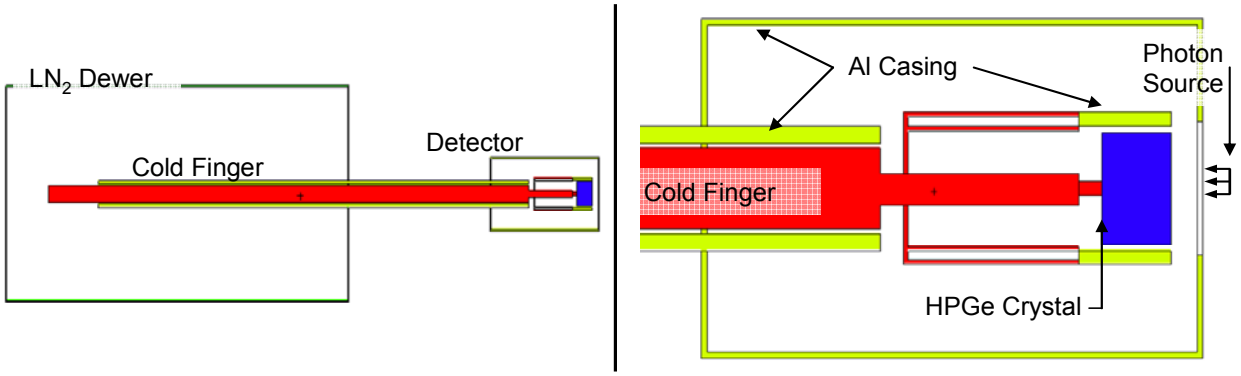

Figure 10: MCNPX for calculating the detector response. The photon tally from the first calculation is transported to the end of the collimator and used as the source. Left: Full geometry. Right: Close up of detector area.

3.4 649C Simulation Results and Comparison

In Figure 11 shows the results for the measurement and corresponding simulation are compared. Immediately it is noticeable that the simulated spectrum is lower in magnitude than the experiment; roughly by a factor of 30. Figure 12 shows the comparison focused on the x-ray region with the simulation results on a separate scale. There are several potential reasons for the discrepancy in magnitude. First, the beta term was ignored; however, the beta term alone cannot be the cause of the discrepancy because in both the simulation and measurement the Pu $K_{\alpha 1}$ x-ray and the ^{155}Eu gamma peaks (~105 keV) are roughly equivalent in magnitude. Given that both gamma and beta sources contribute to the x-ray peaks but only the gamma source contributes to the ^{155}Eu peak, if the ignored beta source term is in fact the dominant contributor, than the ratio of these two peaks would not agree between the simulation and experiment. Second, the ideal collimator assumption used to reduce run times causes lost photon counts. However, if ignoring the reflections inside the collimator was the problem then only the Compton background would be different, the measured and simulated x-ray peak heights would be similar. More probable explanations are that the normalization factor may be inappropriate for the methodology used of breaking the problem into two parts and/or the description of the experimental setup used to create the MCNPX models may be misunderstood, or flawed. For example, the collimator pin hole radius could be mistaken. Small increases in the collimator radius create large increases in the magnitude of the detector response.

A second thing to notice is the large background increase in the 200 keV area, which is significantly more pronounced in the simulation than in the measurement. This is a result of the back scattering off of the cold finger and other metal casings near the detector crystal. The modeled cold finger is based on visual inspection of other HPGe detectors, and has too much heavy material too near to the detector crystal. Fortunately, this effect has a minimal impact on the XRF region.

The comparison of the x-ray region in Figure 12 shows that MCNPX creates x-rays about 0.5 keV higher than seen in the measurement and given in Table 1. This discrepancy is a result of the data libraries used by MCNPX, which have incorrect values for the x-ray peaks tabulated in them. Furthermore, MCNPX incorrectly handles the K_β peaks. All K-shell to M-shell transitions are grouped together and emitted at a single energy, and all K-shell to N-shell transitions are likewise grouped together such that they are all created at the same energy.

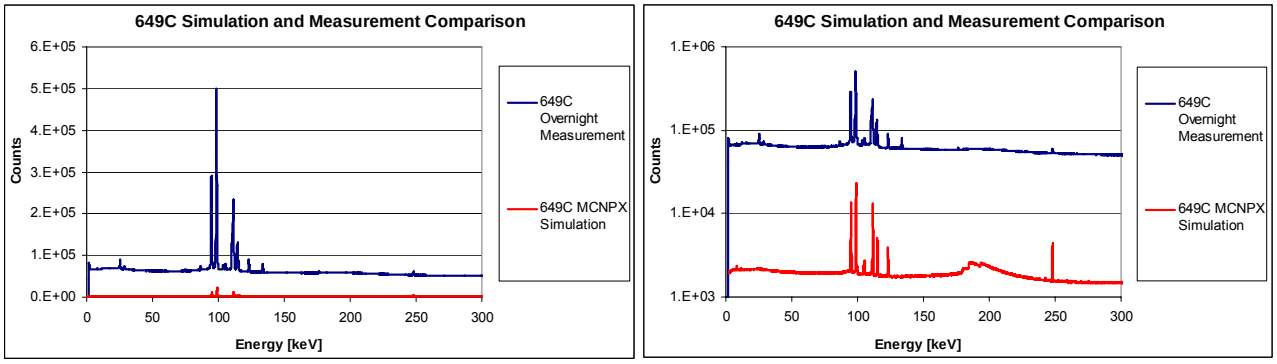

Figure 11: Modeled detector response comparison of the measurement and simulation for pin 649C. Semi-log scale.

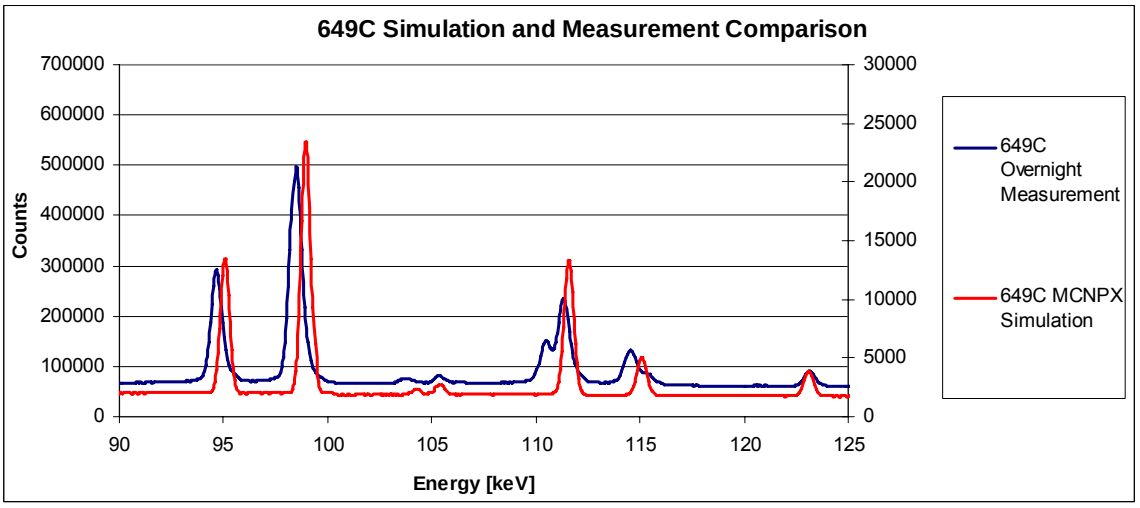

Figure 12: XRF region comparison of measurement and simulation for pin 649C.

3.5 Pu/U Ratio Comparison

The main goal of this work is to test the feasibility of using XRF signals in spent fuel assemblies to quantify elemental Pu through the ratio of Pu/U x-rays. As such, despite the scaling issue stated above, ultimately, the ratio of Pu/U that was simulated is the greatest importance.

To compare the Pu/U ratio between the measured and simulated spectrums, the net peak area of the U $K_{\alpha 1}$ and Pu $K_{\alpha 1}$ x-rays needed to be computed. This was done by averaging several bins on each side of the peak and connecting them linearly to estimate the background under the peak. Then the net peak was calculated by subtracting the estimated background from the total peak counts. Figure 13 and Figure 14 show the U and Pu peaks for both the simulation and measured data. In each case, the different areas marking where the continuum background was averaged along with the estimated peak background is illustrated. The uncertainty for each bin uses counting statistics, where $\sigma_{N,i}$, the uncertainty for bin *i,* is the square root of the number of counts, N_i. No uncertainties for the MCNPX calculation are carried over, and

no uncertainties in the isotopic inventories or source creation are considered. The measured U $K_{\alpha 1}$ and Pu $K_{\alpha 2}$ x-rays are not resolved, such that the Pu $K_{\alpha 2}$ x-ray becomes the tail to the U $K_{\alpha 1}$. From Table 1, the relative intensity of the Pu $K_{\alpha 2}$ x-ray is 62.5% of Pu $K_{\alpha 1}$. Therefore, the expected counts for Pu $K_{\alpha 2}$ can be estimated by multiplying the net counts for Pu $K_{\alpha 1}$ by 0.625. The net counts that should be attributed to just the U $K_{\alpha 1}$ peak are approximated by subtracting out the estimated counts for Pu $K_{\alpha 2}$. For the simulation, the two peaks are resolved enough so that the overlapping was considered negligible.

As discussed earlier, MCNPX produces the U and Pu $K_{\alpha 1}$ x-ray at the incorrect energy. This energy shift becomes a problem when calculating the net counts for Pu because the peak has significant overlap with the ^{155}Eu gamma peak. To avoid this problem, the Pu peak produced by MCNPX in part 1 was shifted down to the correct energy before using the spectrum as the source for part 2. This correction allowed the detected Pu x-ray peaks to be significantly resolved from the ^{155}Eu gamma line such that the overlap is ignored.

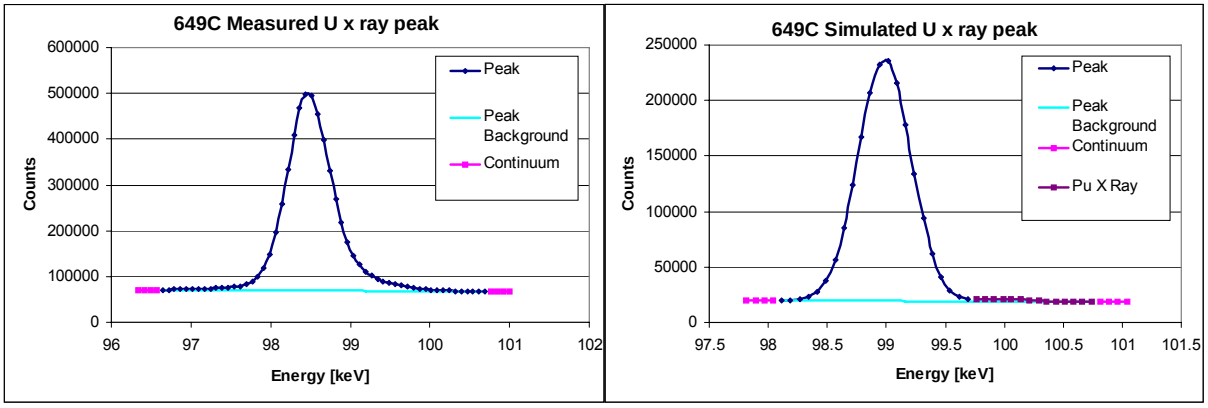

Figure 13: U $K_{\alpha 1}$ peak for 649C. Left: Measured. The tail on the right hand side includes the Pu $K_{\alpha 2}$ peak. Right: Simulated.

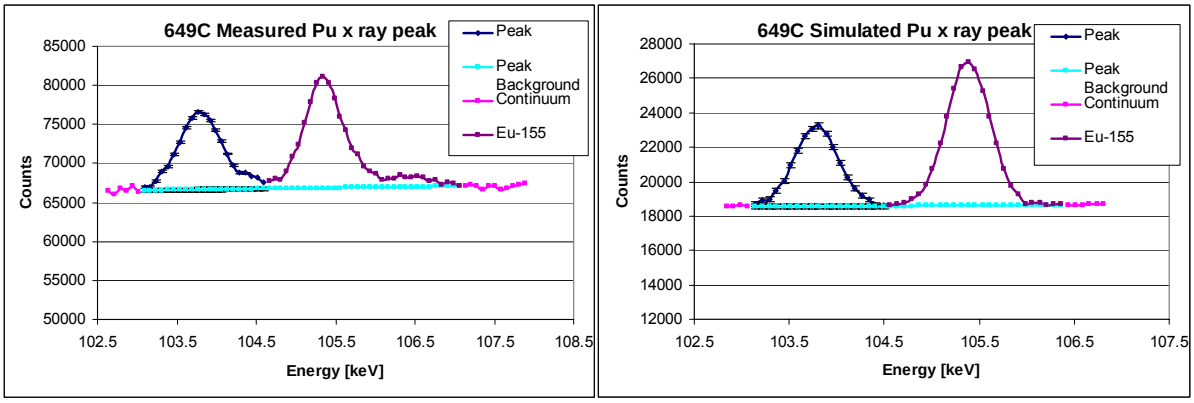

Figure 14: Pu $K_{\alpha 1}$ x-ray for 649C. Left: Measured. Right: Simulated. The energy of the Pu x-ray in the simulation case was corrected in the source term of the MCNPX detector response calculation.

Table 2 gives the results of the peak comparison for pin 649C. The uncertainties are the propagation of counting statistics from each bin. The simulation does an excellent job of calculating the Pu/U ratio, giving the ratio 0.0221±0.0004 compared to the measured ratio of 0.0227±0.0008. The simulated Pu/U ratio differs from the measurement by 2.7%, which is within two sigma of the measured value.

		Total			Peak Background			Net			
U	Measured	7,963,677	+/-	2,822	3,797,929	+/-	1,949	4,107,395	+/-	3,603	(0.1%)
	Simulated	845,405	+/-	919	176,331	+/-	1,956	669,074	+/-	1,011	(0.2%)
Pu	Measured	1,492,617	+/-	1,222	1,399,252	+/-	5,156	93,365	+/-	1,701	(1.8%)
	Simulated	159,962	+/-	400	145,156	+/-	381	14,806	+/-	552	(3.7%)
Pu/U	Measured	0.0227	+/-	0.0004	(1.8%)						
	Simulated	0.0221	+/-	0.0008	(3.7%)						

Table 2: Comparison of Pu and U x-ray peaks for 649C.

There are still many sources of uncertainty that are not accounted for in the above analysis. For example, the inventories calculated for the fuel pin will not be exact. The MCNPX uncertainties for both calculation parts were ignored, and the model itself creates bias because it is a simplification of the measurement setup. Furthermore, the MONTEBURNS/ORIGEN2 depletion code used to calculate the inventories in the model has error that is ignored as well. Destructive analysis is planned for the 649 rod cuts at ORNL in the future; at that point the accuracy of the isotopic inventories can be examined.

Even though the magnitude of the 649C spectrum simulation is wrong, more important is that the 649C pin benchmark does provide confidence that the Pu/U ratio can be predicted. This is an important conclusion since it provides confidence in our ability to make predictions about XRF measurements of assemblies.

4.0 XRF Assembly Models

The suite of MCNPX spent fuel assembly models created for the NGSI effort are used in this work to determine the general behavior of XRF as a function of BU, initial ^{235}U enrichment (IE), cooling time (CT), the overall feasibility, and general limits of detectability for XRF in the context of full assemblies. MCNPX modeling is used to simulate the XRF signal that would be measured with planar HPGe detector for a variety of light-water reactor (LWR) assemblies measured in water. Given that the mean free path of the XRF signal in nuclear fuel is ~0.5mm, obviously this measurement technique is completely blind to scenarios where fuel pins have been removed (that are not in direct field of view). It is assumed that XRF will be used in combination with other techniques more sensitive to fuel pin diversions.

4.1 Spent Fuel Library

The set of spent fuels assemblies created for the NGSI effort used the MCNPX Burnup and Depletion capability to create the isotopic inventories of pins throughout the assembly. The MCNPX Burnup and Depletion capability uses MCNPX to calculate neutron fluxes at power and CINDER'90 to explicitly calculate the subsequent buildup and decay of isotopic inventories for each time step. While CINDER'90 uses a data set of 3400 nuclides, MCNPX discards all but the few hundred isotopes that have neutron cross sections available for calculating flux. It is assumed that the isotopes tracked are significant for our purposes. The array of assemblies produced is referred to as the spent fuel library (SFL) and consists of the 64 combinations of the following: 15, 30, 45, and 60 Gwd/tU BU; 2, 3, 4, and 5% IE; and 1, 5, 20, and 80 years CT.

Memory allocation limitations with the MCNPX code used to produce the SFL confines the number of BU materials to a few hundred. Restricting the number of radial regions down to four for each pin, results in a loss of fidelity in modeling the radial Pu. Furthermore, only one vertical region was modeled, thus, there is no vertical BU profile in the SFL used in this work.

The assemblies created for the SFL used a 17 by 17 Westinghouse LWR assembly with infinitely reflected boundaries. This boundary condition is good for creating generic assemblies; however, it neglects asymmetries in BU throughout the core that arise from fuel loading schemes. Asymmetries in BU, and therefore Pu, may be particularly difficult for the XRF technique to handle because the measured U and Pu x-rays can only be sampled from the front row of pins. Extrapolating the Pu content of the whole assembly from the measured outer pins will be complicated by these asymmetries.

Recent improvements in the MCNPX code have alleviated memory restrictions to some degree, increasing the potential number of materials that can be burned [11]. Combined with increases in computational resources, the MCNPX code improvements have allowed a current effort to create a new SFL. This second SFL is being created with a $1/8^{th}$ core model so that neighboring assemblies can be on different cycles. Assemblies created in this new SFL will have a better representation of asymmetries in BU resulting from reactor loading schemes [12]. Exploring the effect of BU asymmetry in the assembly is a topic for future work in XRF.

4.2 Modeling Methodology

The methodology for simulating detector response to spent fuel assemblies is similar to the methodology employed for simulating pin 649C in that the MCNPX component is broken down into two parts. The first step is to calculate the initial source, then use MCNPX to calculate the photon spectrum that escapes one face of the assembly. Next, the magnitude is corrected for geometry of the collimator, and finally, another MCNPX simulation is used to calculate the detector response, using the energy spectrum from the first MCNPX calculation as the source for the second. Figure 15 illustrates the methodology.

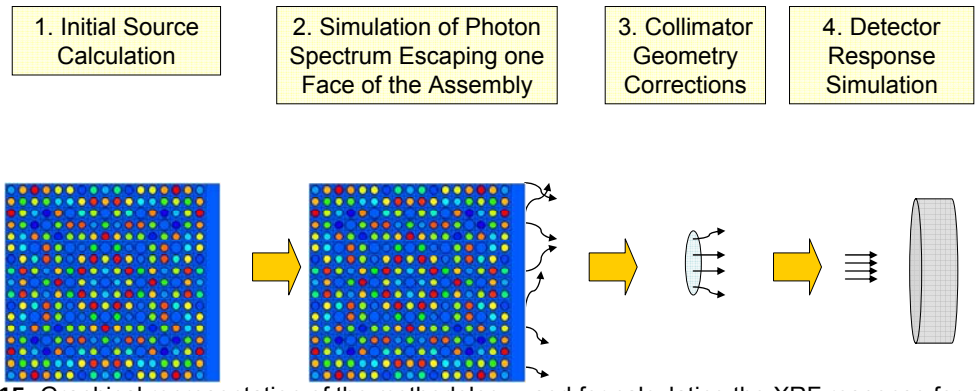

Figure 15: Graphical representation of the methodology used for calculating the XRF response for an entire assembly.

Using the isotopic inventories created for the SFL described above, the same code used to calculate the gamma source for the 649C benchmark is used to create the gamma source definition for the entire spent fuel assembly. Only the gamma source is considered. In the 649C pin simulations, the beta source was estimated to contribute 10% to the XRF signal.

There is also reason to believe beta particles will be less important for assembly measurements than single pins. High energy photons in inner pins can penetrate to the outer pins where fluorescence of U and Pu can contribute to the x ray signal. Betas born in inner pins deposit their energy locally and only some fraction will produce a high energy gamma as a secondary particle to transport their energy through the assembly to the edge.

The assemblies are 21.42 cm to a side and only 6 cm of rod height is modeled. There is a possibility that structural objects or operator regulations may prevent a collimator from being placed directly against the side of the assembly. Therefore, 1.5 cm of water is modeled on the side of the assembly that is tallied. In MCNPX, a surface tally with energy bins of 0.1 keV is used to calculate the spectrum that escapes the side of the assembly being measured. Since the SFL has symmetrical BU on each side, there is no difference on which side is tallied. Similar to the simulation of 649C, a maximum angle of 20º to the surface normal is allowed in the tally. Figure 16 shows the MCNPX model used for comparing the Pu/U ratios of the SFL.

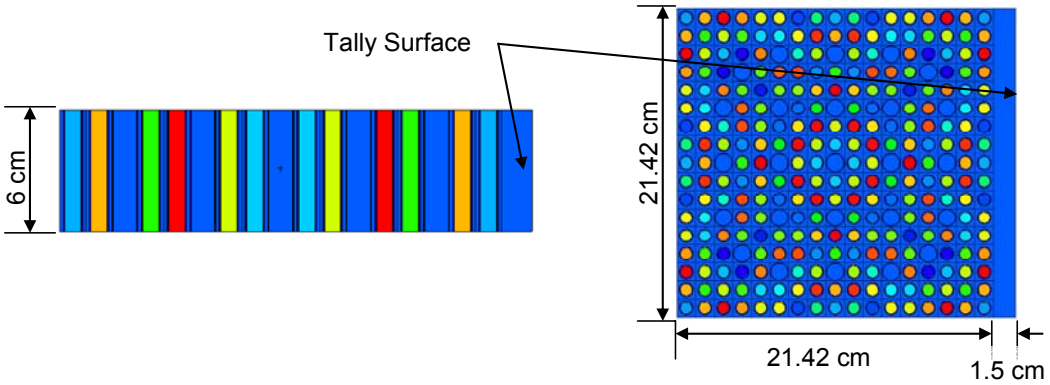

Figure 16: MCNPX model of spent fuel assembly. One side has 1.5 cm of water and is tallied on.

It is important to note that with this modeling setup, one whole side of the assembly is tallied together. Therefore, spatial information from a single pin or section is effectively lost. Our current computational resources force the use of this technique. However, given that the desired quantity is Pu in an assembly, measuring multiple pins will be necessary in any event. Thus, taking the tally of an entire side will be relatable to the measurement sum of each pin along one face of the assembly.

A collimator that receives photons from an entire assembly side will likely have too high of a count rate or too narrow a slit to be practical given the need to keep the count rate low. Using a pinhole collimator is more realistic. Scanning across the side of an assembly can keep count rates at acceptable levels while measuring more than one pin. However, using multiple detectors is more likely as a means of determining the Pu mass across the face of the assembly as well as reducing the overall count time.

Unlike the simulations of 649C, there is no pre set collimator length or diameter. Therefore, one can be chosen that optimizes the count rate experienced by the detector. Given that the source strength of the assemblies tested varies drastically - consider 1 and 80 years CT - it will be beneficial to have several different collimator sizes. The magnitude of the detector response tallied in the second MCNPX calculation will need to be normalized to the gamma source strength of the assembly and the collimator used. Unlike the 649C benchmark, the correction for the collimator will be calculated by considering two basic components of the MCNPX tally and the collimator. The benefit of calculating correction factors for the collimator over using MCNPX as in the 649C benchmark is that count rates in the detector can be adjusted in post processing, instead of requiring multiple MCNPX runs for different collimator dimensions. The first step is a correction factor for the difference in area that is tallied, $A_{TALLY,}$ and the cross sectional area of the collimator, A_{TRUE}. This correction factor, f_{Area}, is calculated with the equation below.

$$f_{AREA} = \frac{A_{TRUE}}{A_{TALLY}}$$

The second correction factor takes into account the fact that the MCNPX tallies all photons that escape the tally surface at an angle below 20º to the surface normal. The angular correction factor, f_θ, is calculated by taking the ratio of steradians, Ω, between the maximum angle that incident photons can reach the end of the collimator with, θ_{TRUE}, and the maximum angled tallied, θ_{TALLY}, collimator and what was tallied.

$$f_\theta = \frac{\Omega_{TRUE}}{\Omega_{TALLY}} = \frac{1-\cos\theta_{TRUE}}{1-\cos\theta_{TALLY}}$$

The normalization constant applied to the MCNPX detector response spectrum is the product of the total source strength of gammas over the measurement time, f_{Area}, and f_θ. This approach implies an ideal

collimator, as well as a collimator hole under vacuum. For measurement purposes the collimator hole will need to be sealed from water to avoid complete attenuation of the x ray signal.

Tolerable count rates of $3 \cdot 10^4$ to $5 \cdot 10^4$ Hz can be expected from commercial equipment (although 10^5 Hz is available as well). In this work, the modeled collimator dimensions are chosen so that the total count rate experienced by the detector - including events that are rejected by the lower level discriminator and events that are above the gain settings - stay in that range. Since the magnitude of the 649C benchmarking was off by an order of magnitude and the normalization uses a different method, there is no reason to expect the assembly modeling not also be off in magnitude. This is of little consequence since the magnitude of the detector response can be adjusted in measurements by adjustment in collimator aperture and the large variation of source intensities will already require a set of different sized collimators. More importantly, regardless of how different the collimator modeled and the actual collimator end up being, as long as the count rate experienced by the actual detector falls into the range used as the criterion for choosing the modeled collimator, then the live-times and counting statistics presented in the following work will still be accurate.

4.3 Signal Contribution by Assembly Row

One of the limiting factors in application of XRF is the lack of penetrability of a 100 keV photon through fuel. The mean free path of x-ray energy photons in nuclear fuel is about 0.05 cm. Thus, x-rays created in the second row of pins have a negligible chance of crossing through a front row pin to reach a collimator or detector. This problem is compounded by the high count rate which limits collimation to one pin and potentially by the high energy photons emitted by fission products deeper in the spent fuel assembly, which can penetrate through several rows of fuel. Therefore, pins not in the front row can contribute to Compton background with high energy gammas, but their x-rays never make it to the detector.

Using an assembly with 60 GWd/tU BU, 4% IE, and 5 year CT, the detector response to each row in the assembly is simulated. A collimator 80 cm long with a radius of 0.15 cm and a live-time of 3600 seconds is assumed. The total count rate calculated on the detector is about $4.6 \cdot 10^4$ Hz. Figure 17 shows the contribution to the signal from each row for the U $K_{\alpha 1}$ energy region and Figure 18 plots the percentage of net peak and background for each row. Both the x-ray signal and background contributions drop off very rapidly in the first five rows. By the ninth row, the contribution to net peak signal is down to 0.1%, and the background is 0.6%. Effectively, the second half of the assembly does not significantly contribute to the signal or background. Rows 9 through 17 combine to contribute less than 0.5% to the net peak and 2% to the background. An important point to note is that the rows beyond the first row do contribute to XRF signal. X-rays created outside of the first row don't contribute to the signal, but the higher energy gamma field that penetrates to the first row does create x-rays in the first row that do contribute to signal. This allows the x-ray signal to grow almost proportionately with the Compton background, which means that the additional rows of pins in an assembly won't necessarily mask the Pu x-ray signal.

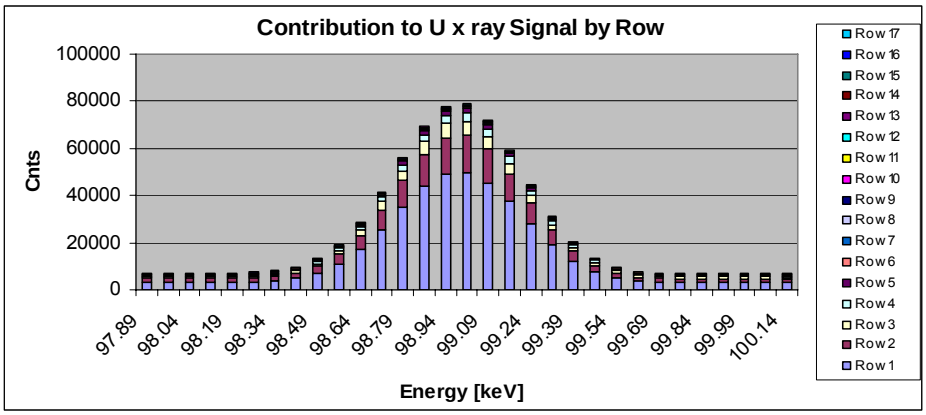

Figure 17: Contribution of each row to the total peak measured. 60 Gwd/tU BU, 4% IE, and 5 year CT, with a 3600 s live-time.

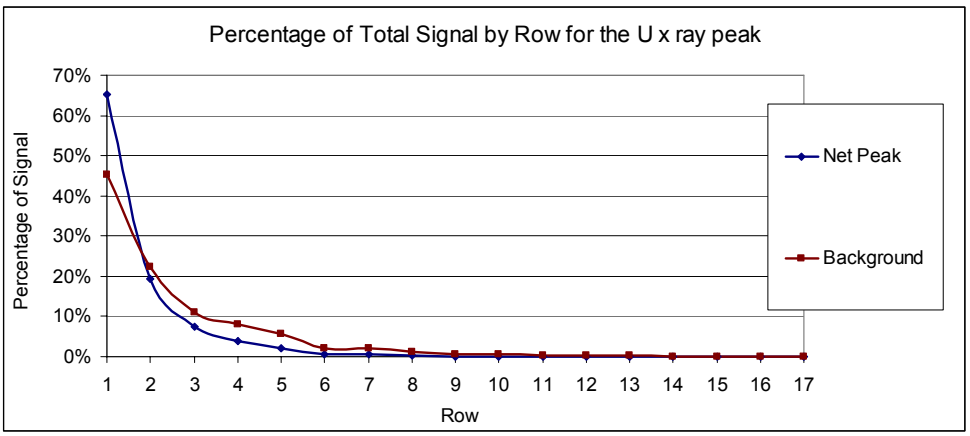

Figure 18: The contribution of each row to the detected signal and background for the U $K_{\alpha 1}$ peak.

4.4 Detector Crystal Thickness

To minimize count time, it is desirable to choose a detector crystal that provides the highest signal-to-background for the x-ray peaks of interest. For detector optimization, the 45 Gwd/tU BU, 4% IE, and 5 year CT assembly was used. MCNPX simulations using the model seen in Figure 10, except with varying crystal thicknesses, were run. Thickness ranged from 1.5 cm, like the crystal used in 649C, down to 0.1 cm. A collimator 80 cm long with a diameter of 0.15 cm is assumed. The count rate experienced by the different detectors ranged from 3.2 to $4.4 \cdot 10^4$ Hz. Figure 19 shows the peak with uncertainties for counting statistics assuming a 1 hour count time, and Figure 20 shows the calculated signal to background ratio for the Pu x-ray peak as a function of detector thickness. The Pu $K_{\alpha 1}$ x-ray is the peak examined for comparison because its smaller signal makes it the limiting factor in measuring the Pu/U ratio accurately. A detector crystal with a thickness between 0.25 cm and 0.75 cm should be optimal for measuring XRF of spent fuel assemblies. In the remaining work, the HPGe crystal thickness used is 0.5 cm.

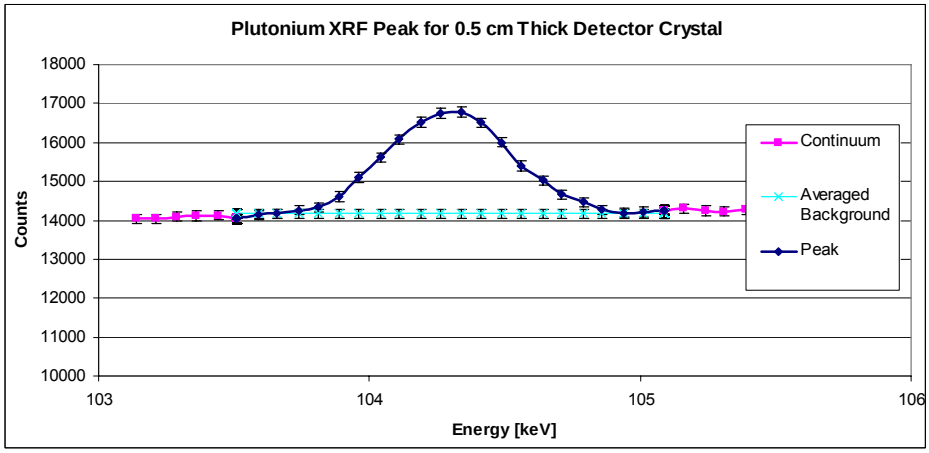

Figure 19: Pu x-ray peak for a 0.5 cm thick HPGe crystal showing the peak, continuum, and peak background. Counting statistics are for 1 hour live-time. Assembly modeled is 60 Gwd/tU BU, 4% IE, and 5 year CT.

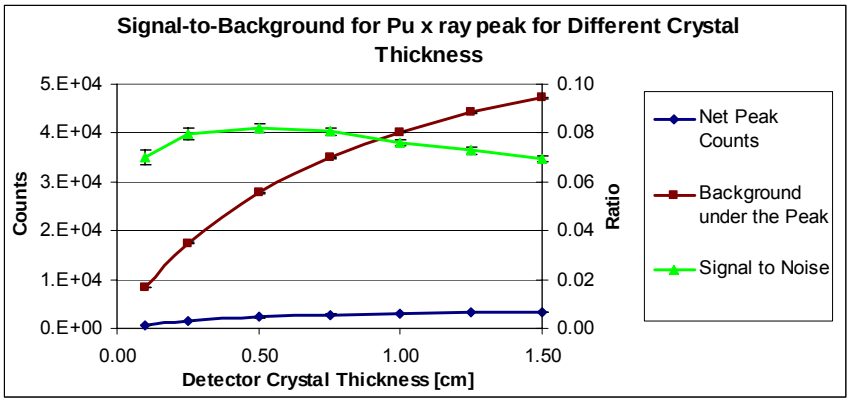

Figure 20: Signal-to-background ratio as a function of HPGe crystal thickness for the Pu x-ray peak. Counting statistics are for 1 hour live-time.

4.5 Detector Crystal Radius

This section examines the signal-to-background for x-rays of interest for various crystal radii. In the 649C measurements, a crystal radius of 1.26 cm was used. The collimator radius was only 0.1 cm. Therefore, much of the crystal was unused. Radii simulated ranged from 1.26 cm, like the crystal used in 649C, down to 0.25 cm. Figure 21 shows the calculated signal to background ratio for the Pu x-ray peak as a function of HPGe crystal radius. These simulations suggest that the effects of radius are not as important as crystal thickness. This is expected since the photon beam is directed at the entire thickness of the crystal, but the collimator forces the beam to be directed at only 0.15 cm of radius. Since the collimator radius for measuring assemblies is not set, a good choice of crystal radius may be at least twice the radius of the collimator. The remaining models use a 0.5 cm radius HPGe crystal.

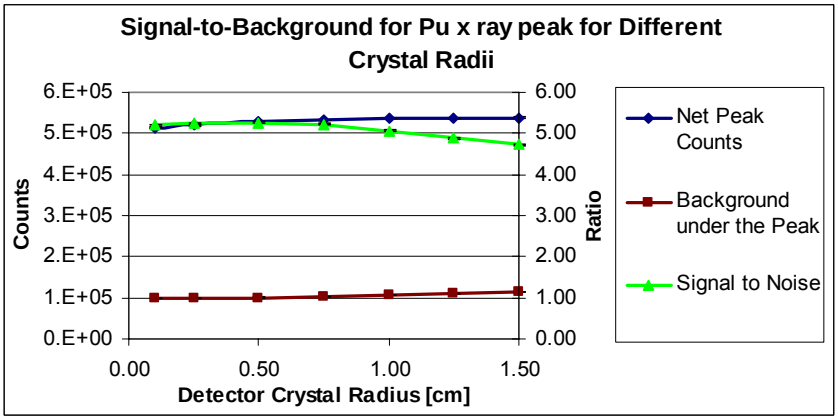

Figure 21: Signal-to-background ratio as a function of HPGe crystal radius for the Pu x-ray peak. Counting statistics are for 1 hour live-time.

4.6 Water Thickness

Different conditions among facilities make the amount of water that will be present between the assembly and collimator an unknown variable. The mean free path of 100 keV photons in water is approximately 6 cm; therefore, small amounts of additional water are not expected to impair XRF. Thicknesses of water were simulated ranging from 0 to 2 cm. Figure 22 shows the calculated signal-to-background ratio for the Pu x-ray peak as a function of water thickness. Having a collimator 2.0 cm away from the assembly is expected to drop the signal-to-background roughly 30% below the case where the collimator is touching

the assembly. The assembly cases discussed later comparing BU, IE, and CT, all assume that 1.5 cm of water is between the assembly and collimator.

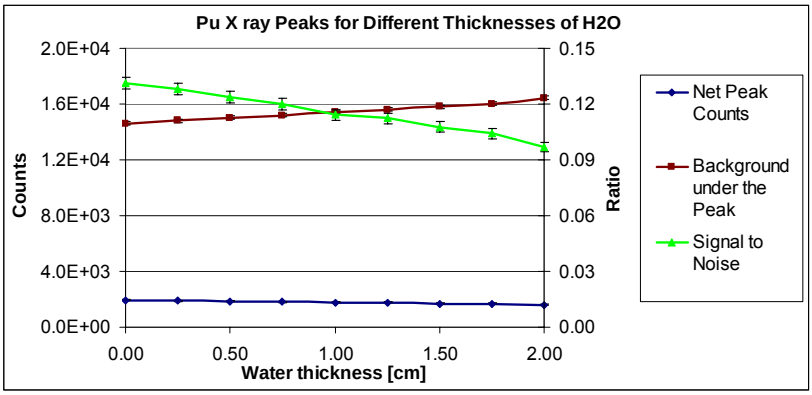

Figure 22: Signal-to-background ratio as a function of H_2O thickness between the assembly and collimator. Counting statistics are for 1 hour live-`time.

4.7 Pu/U Ratio of Spent Fuel Assemblies

Using the methodology described above, the suite of SF assemblies in the SFL was simulated using an HPGe crystal that was 0.5 cm thick with a 0.5 cm radius. The collimator dimensions needed to be varied to adjust for the wide range of photon source strengths. For example, the photon source strength of assemblies with 80 year CT typically is ~20 times weaker than the 1 year CT scenario. Assuming that five collimators, 80 cm long with radii of 0.3, 0.25, 0.2, 0.15, and 0.1 cm, are available, for each assembly the biggest collimator was chosen such that the count rate experienced by the detector was less than $4 \cdot 10^4$ Hz. The Pu/U XRF ratios calculated compared to the Pu/U mass ratios for the SFL is plotted in Figure 23 grouped by BU (color) and IE (shape). CT has a small impact on the Pu/U ratio, the exception being 80 years CT, where the 14 year half-life of ^{241}Pu and 87.7 year half-life of ^{238}Pu can reduce the Pu mass significantly. CT does significantly affect the total source strength. As expected, BU has a large impact on Pu and thus, the Pu/U ratio. The 15 Gwd/tU BUs are grouped together with much lower Pu/U ratio than the rest.

The uncertainties in the inventory calculation as well as the MCNPX models are completely ignored in the results. Since the problem is broken into two parts, the uncertainties in the first part are lost by default. The uncertainties presented are from counting statistics for a 1 hour live-time, and are unrelated to all of the modeling uncertainties that were ignored. Hence, this is why none of the 64 data points in Figure 23 are outside 1 sigma from the linear relationship.

The linear fit for the data shows a very strong correlation with an R^2 value of 0.9899. This suggests that if counting statistics can be driven down by an appropriate collimator and count time, XRF may be used to accurately determine the Pu quantity. However, it has been demonstrated that the Pu/U XRF signal from the assembly is really a representation of the Pu/U mass ratio in the front row of pins. The reason it matches so well to the Pu/U mass ratio of the assembly is due to the material symmetry of the assembly caused by assuming infinite boundaries. Depletion calculations which use spent fuel assemblies with a more realistic fuel loading, and therefore asymmetries in the Pu distribution, need to be modeled to determine a more accurate representation of how the Pu/U XRF signal will compare to the Pu/U mass in an assembly.

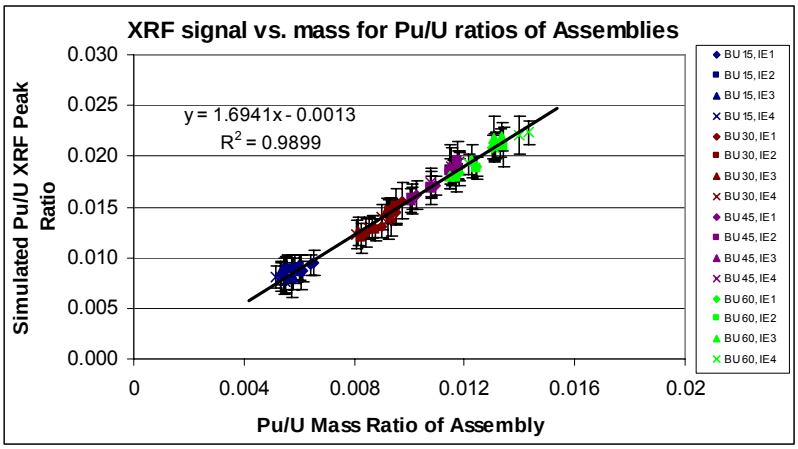

Figure 23: Simulated Pu/U x-ray ratio compared to the Pu/U mass ratio of the assembly. All modeling uncertainties are ignored. Uncertainties presented are from counting statistics assuming a 1 hour live-time using various sized collimators. Hence, none of the 64 data points appear outside 1 sigma from the linear relationship.

Table 3 lists the Pu/U mass ratio, Pu/U XRF simulation ratio for the 64 assemblies modeled, the collimator radius used, along with the XRF simulation uncertainty from counting statistics for a 1 hour and 10 hour measurement. For a 1 hour count time, the highest relative uncertainty for the XRF Pu/U ratio was 19.1%, which corresponds to an uncertainty on the Pu/U mass ratio of 15.5%. Excluding the low Pu/U ratios of the 15 Gwd/tU BU cases, the highest uncertainty on the XRF Pu/U ratio was 14.1%, corresponding to 12.5% uncertainty for the Pu/U mass ratio. For the 10 hour count time the highest uncertainty was 6.0% for the XRF Pu/U simulation, which corresponds to 3.6% uncertainty for the Pu/U ratio of the mass in the SF assembly. The correlation for uncertainty is found by using the correlation from the linear fit in Figure 23.

For reasonable results with a single detector and a well chosen collimator diameter, a count time of at least 10 hours should be expected. This will not be acceptable for a technique that needs to quickly assay hundreds of assemblies. One solution is to have an array of collimators and detectors collecting and combining signals from an assembly.

Burnup	Initial ^{235}U Enrichment	Cooling Time	Pu/U Ratio		Collimator Radius	Uncertanty From Counting Statistics	
			Mass From SF Library	XRF Measurement			
[GWd/MtU]	[%]	[Years]			[cm]	1 hour	10 hours
15	2	1	0.0065	0.0095	0.15	13.7%	4.3%
		5	0.0064	0.0093	0.2	11.2%	3.5%
		20	0.0061	0.0087	0.25	11.3%	3.6%
		80	0.0059	0.0088	0.3	15.4%	4.9%
	3	1	0.0061	0.0085	0.15	16.0%	5.1%
		5	0.0060	0.0090	0.2	11.8%	3.7%
		20	0.0058	0.0089	0.25	11.0%	3.5%
		80	0.0056	0.0088	0.3	15.5%	4.9%
	4	1	0.0058	0.0082	0.15	17.3%	5.5%
		5	0.0057	0.0081	0.2	13.4%	4.2%
		20	0.0055	0.0084	0.25	11.6%	3.7%
		80	0.0054	0.0085	0.3	16.1%	5.1%
	5	1	0.0055	0.0076	0.15	19.1%	6.0%
		5	0.0055	0.0081	0.2	13.5%	4.3%
		20	0.0053	0.0081	0.25	12.1%	3.8%
		80	0.0052	0.0081	0.3	16.9%	5.3%
30	2	1	0.0098	0.0156	0.1	11.0%	3.5%
		5	0.0095	0.0145	0.15	8.1%	2.6%
		20	0.0090	0.0130	0.2	8.4%	2.6%
		80	0.0084	0.0124	0.25	11.4%	3.6%
	3	1	0.0095	0.0150	0.1	12.0%	3.8%
		5	0.0093	0.0146	0.15	8.3%	2.6%
		20	0.0088	0.0127	0.2	8.6%	2.7%
		80	0.0082	0.0119	0.25	11.8%	3.7%
	4	1	0.0094	0.0140	0.1	13.4%	4.3%
		5	0.0092	0.0146	0.15	8.5%	2.7%
		20	0.0087	0.0131	0.2	8.4%	2.6%
		80	0.0082	0.0126	0.25	11.2%	3.5%
	5	1	0.0092	0.0137	0.1	14.1%	4.5%
		5	0.0090	0.0140	0.15	9.1%	2.9%
		20	0.0086	0.0129	0.2	8.5%	2.7%
		80	0.0081	0.0122	0.25	11.6%	3.7%
45	2	1	0.0118	0.0190	0.1	6.8%	2.2%
		5	0.0116	0.0184	0.1	11.3%	3.6%
		20	0.0109	0.0171	0.15	9.4%	3.0%
		80	0.0103	0.0162	0.25	7.2%	2.3%
	3	1	0.0118	0.0195	0.1	6.9%	2.2%
		5	0.0115	0.0185	0.1	11.5%	3.6%
		20	0.0108	0.0169	0.15	9.5%	3.0%
		80	0.0101	0.0158	0.25	7.4%	2.3%
	4	1	0.0118	0.0191	0.1	7.3%	2.3%
		5	0.0115	0.0189	0.1	11.5%	3.6%
		20	0.0108	0.0169	0.15	9.5%	3.0%
		80	0.0100	0.0156	0.25	7.5%	2.4%
	5	1	0.0118	0.0194	0.1	7.5%	2.4%
		5	0.0115	0.0190	0.1	11.7%	3.7%
		20	0.0108	0.0174	0.15	9.3%	2.9%
		80	0.0101	0.0158	0.25	7.4%	2.3%
60	2	1	0.0133	0.0213	0.1	5.2%	1.6%
		5	0.0130	0.0209	0.1	8.6%	2.7%
		20	0.0124	0.0189	0.15	7.5%	2.4%
		80	0.0118	0.0184	0.25	5.6%	1.8%
	3	1	0.0134	0.0209	0.1	5.3%	1.7%
		5	0.0131	0.0211	0.1	8.5%	2.7%
		20	0.0124	0.0188	0.15	7.5%	2.4%
		80	0.0117	0.0179	0.25	5.7%	1.8%
	4	1	0.0134	0.0219	0.1	5.3%	1.7%
		5	0.0131	0.0217	0.1	8.5%	2.7%
		20	0.0123	0.0195	0.15	7.3%	2.3%
		80	0.0115	0.0180	0.25	5.7%	1.8%
	5	1	0.0144	0.0223	0.1	5.3%	1.7%
		5	0.0140	0.0221	0.1	8.5%	2.7%
		20	0.0131	0.0208	0.15	6.8%	2.2%
		80	0.0121	0.0192	0.25	5.3%	1.7%

Table 3: List of Pu/U mass ratio, Pu/U XRF simulation ratio, collimator radius, and Pu/U uncertainty from counting statistics for both 1 hour and 10 hour count times for the suite of assemblies modeled.

The uncertainty in Table 3 for a single detector is determined by (1) the relative intensity of the peak to the continuum, which was modeled reasonably well in the 649C benchmark, and (2) the count rate limit of the detector system is adjusted to a near optimum value by increasing the collimator radius. Given the small spot size, this adjustment keeps the observed area limited to one pin. The conclusion can then be drawn that the uncertainty of a single detector cannot be improved above what is listed in Table 3. If the simulations for an assembly are off by an order of magnitude, as was the case for the pin result, one would simply reduce the collimator area by that factor and get the same uncertainty for a 1 hour count.

As a result, it can be concluded that a single detector XRF system would not be practical for a spot inspection systems at pool storage facilities since the uncertainty from count statistics after a 1 hour count would be between 7% and 12% (30 GWd/tU and 45 GWd/tU for 1 to 80 year cooling times). This count time is long for spot inspections and no correction has been made for extrapolating from a few to several mm^2 observation area on the edge to fuel to the total Pu in the assembly. A point for future research is how many detector crystals could be combined in a given volume, weight, cost system to make a practical system for spot inspection, depending on if it is shown to be feasible to extrapolate from the Pu determined on the edge of the assembly to the whole assembly. More specifically, it would be desirable to have a system that obtains uncertainty of ~2% in 1000s. Based on the results in Table 3, a reasonable estimate is that ~100 detectors will needed.

The next step is to think how an XRF system comprised of an array of detectors could meet the two other safeguard needs we are focused on. It is expected that an XRF system could be retrofitted into a reprocessing facility, and it certainly could be designed into any future reprocessing facility. If it is shown to be feasible to extrapolate from the Pu determined on the edge of the assembly to the whole assembly, then a point of necessary future research is to determine how many detectors can be practically positioned around the assembly. It is anticipated that, with enough funding, the uncertainty could be reduced to below 2%. Furthermore, an array of detectors around the assembly would be a necessary step in doing the best possible extrapolation from the edge measurement of XRF to the entire assembly.

Given the low energy of the x-rays relative to the rest of the spectrum coming from spent fuel, essentially the only way to reduce the count rate to a level that the detection system can handle is to collimate. Scanning the collimator values in Table 3 we see that the values range from 1 to 3 mm with the vast majority of the more realistic inspection cases having a radius of 2 mm or less; the 1 mm radius would result in a ~3 mm^2 projected collection area on the fuel. Therefore, combining the observed surface area with the fact that the mean free path of a 100 keV photon is around 0.5 mm, we can conclude that the majority of the signal obtained from a single detector in an XRF system will obtain its signal from a volume in the fuel of 1 to 20 mm^3. This is an important number since it quantifies the task of extrapolating from edge measurements to the whole of the assembly.

4.8 Pu Profile

Symmetry from the infinite boundary condition used in the SFL creation is partially responsible for the excellent agreement between the simulated XRF Pu/U ratios and the Pu/U mass calculated in the assembly. Preliminary work in creating the second SFL mentioned previously was used to produce Figure 24, which gives the Pu mass in the outer ring of pins for a 45 Gwd/tU BU, 4% IE, and 5 year CT. For this example, the Pu mass on one face of the assembly is about 10% less than another face. The effects of asymmetric Pu distribution and the general problem of Pu extrapolation from the measured front pins is left for future work.

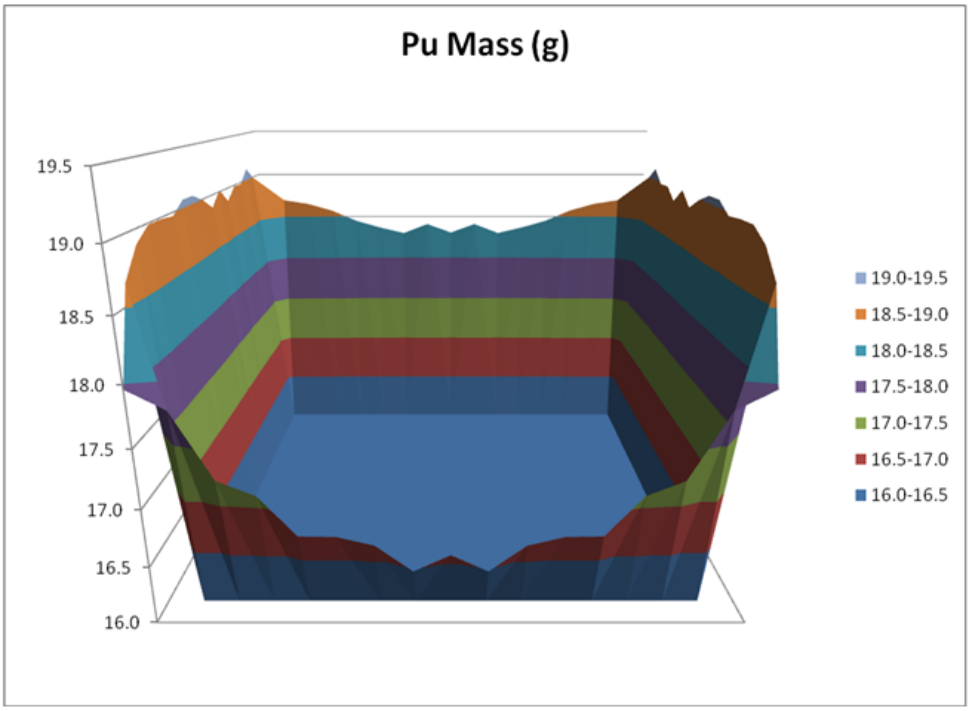

Figure 24: The Pu mass in the outer ring of pins for an assembly with 45 Gwd/tU BU, 4% IE, and 5 year CT.

5. Conclusion

An XRF measurement of a spent fuel rod section, labeled 649C, made at ORNL was simulated to determine our capability to model XRF signals. Modeling of XRF signatures in spent fuel with MCNPX presents many challenge because of the heavy collimation needed to suppress the high photon field. Breaking the problem into two separate MCNPX calculations made the simulation possible with available resources. Modeling of the 649C fuel pin suggest that the beta emission in spent fuel accounts for roughly 10% of the XRF signal. The magnitude of the 649C simulation spectrum was off by an order of magnitude; however, the Pu/U ratio simulated is in good agreement with the Pu/U ratio measured; suggesting a normalization problem. The Pu/U ratio simulated differed from the measurement by 2.65%, which is within 3 sigmas of the measured data. Successful modeling of the Pu/U ratio for a single fuel pin, gave us confidence in our ability to model XRF signals from spent fuel.

Modeling of the spent fuel library indicated that despite the low penetrability of x-rays in spent fuel, it should be possible to observe measurable Pu x-ray peaks. Furthermore, across the suite of different BUs, IEs, and CTs simulated, the XRF Pu/U ratio had a very strong correlation with the Pu/U mass ratio present in the fuel, suggesting that XRF is a feasible technique for quantifying Pu in spent nuclear fuel. Uncertainty in the Pu/U ratio from counting statistics varied greatly between assemblies because of the variation in the gamma intensities that primarily drive the XRF signal. Choosing the collimator radius to correct for different source intensities, a single detector with a 1 hour count time can expect approximately 10% uncertainty. A 10 hour count time corresponded to roughly a 4% uncertainty in the Pu/U ratio.

The spent fuel assemblies used in the simulation were created using an assumption of symmetry that is not representative of real world reactor loading plans, where different sides of an assembly can have very different BUs, and thus, different Pu masses. Given that the low penetrability of x-rays in fuel means that XRF is only measuring the outermost pin in an assembly, the ability with which one can accurately extrapolate the Pu profile from the edge to the entire assembly is the key issue in assessing the utility of XRFs for quantifying Pu mass in a full assembly. Future work with assemblies that do not have symmetrical BUs is needed to examine the effect this will have on XRFs feasibility as an assay technique for spent fuel assemblies.

6. Acknowledgments

The authors would like to acknowledge the support of the Next Generation Safeguards Initiative Office of Nuclear Safeguards & Security (NA-241) National Nuclear Security Administration U.S. Department of Energy

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The Role of Monte Carlo Burnup Calculations in Quantifying Plutonium Mass in Spent Fuel Assemblies with Non-Destructive Assay

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Abstract:

The Next Generation Safeguards Initiate (NGSI) of the United States Department of Energy has funded a multi-laboratory/university collaboration to quantify plutonium content in spent fuel (SF) with non-destructive assay (NDA) techniques and quantify the capability of these NDA techniques to detect pin diversions from SF assemblies. The first Monte Carlo based spent fuel library (SFL) developed for the NGSI program contained information for 64 different types of SF assemblies (four initial enrichments, burnups, and cooling times). The maximum amount of fission products allowed to still model a 17x17 Westinghouse pressurized water reactor (PWR) fuel assembly with four regions per fuel pin was modelled. The number of fission products tracked was limited by the available memory. Studies have since indicated that additional fission product inclusion and asymmetric burning of the assembly is desired. Thus, an updated SFL has been developed using the Monte Carlo-based burnup code *Monteburns*, which links MCNPX to a depletion code and models a representative 1/8 core geometry containing one region per fuel pin in the assemblies of interest, including a majority of the fission products with available cross sections.

Often in safeguards, the limiting factor in the accuracy of NDA instruments is the quality of the working standard used in calibration. In the case of SF this is anticipated to also be true, particularly for several of the neutron techniques. The fissile isotopes of interest are co-mingled with neutron absorbers that alter the measured count rate. This paper will quantify how well working standards can be generated for PWR spent fuel assemblies and also describe the spatial plutonium distribution across an assembly. More specifically we will demonstrate how Monte Carlo gamma measurement simulations and a Monte Carlo burnup code can be used to characterize the emitted gamma spectrum and the asymmetries experienced in the second SFL.

Keywords: spent fuel, plutonium distribution, nuclear safeguards, non-destructive assay

1. Introduction

According to the Information Circular (INFCIRC) 153[1], the technical objective of International Nuclear Safeguards is "… the timely detection of diversion of significant quantities of nuclear material from peaceful nuclear activities … and deterrence of such diversion by risk of early detection". In support of this objective a five year research effort was started in March, 2009, by the Next Generation Safeguard Initiative (NGSI) of the U.S. Department of Energy[2]. Initial efforts have been invested in Monte Carlo simulations of various detector designs. One item of great importance to the accurate assessment of the effectiveness of a particular detector design is the spent fuel composition in the fuel assembly being analyzed. The first phase of spent nuclear fuel modelling in support of the NGSI effort included significant effort by Fensin et al in the creation of Spent Fuel Library number 1 (SFL1)[3] using the MCNPX in-line burnup (BU) capabilities[4]. The simulation was performed using an infinitely reflected generic 17x17 PWR fuel bundle, utilizing 1/8 assembly symmetry. In an effort to more accurately capture the asymmetric spectral effects resulting from a fuel shuffling sequence, a second spent fuel library (SFL 2a)[5] has been developed which utilizes increased computational capabilities

coupled with new updates in MCNPX 2.7.d2 reducing memory requirements[6], allowing more realistic core shuffling sequences to be modeled. Using SFL 2a and two alternate shuffling sequences, pertaining to SFL 2b and 2c, which were simulated to provide additional data points for the assessment of spatial dependencies, spatial plutonium distribution and the dependence upon fuel shuffling schemes (core loading patterns) was investigated.

In addition to efforts invested in the characterization of plutonium in SFLs 2a, 2b and 2c, the asymmetric BU distribution also presented a more realistic starting point for performing passive gamma simulations in support of average BU estimation. Numerous studies have been performed investigating the accuracy of passive gamma measurements for BU determination including work by Hsue et al[7], Tsao and Pan[8], Fensin et al[9][12][13], and Phillips and Bosler[11]. In this study, the viability of coupled Monte Carlo based BU calculations with MCNPX detector simulations as applied to assemblies containing asymmetric spatial BU distributions is investigated.

2. Spent Fuel Library #2

The first spent fuel library created in support of the NGSI effort included a fully populated matrix consisting of four initial enrichments (IE) at 2, 3, 4 and 5%, four BU values of 15, 30, 45 and 60 GWd/MTU, and four cooling times (CT) of 1, 5, 20 and 80 years. In the creation of the second spent fuel library some data points were removed since they represented an unlikely domain space in typical reactor operation. The high BU, low IE data points, and all associated CTs of 2, 3% IE crossed with 45, 60 GWd/MTU were removed, as well as the 4% IE, 60 GWd/MTU data point. In place of these removed data points, the number of CT included with the remaining cases was increased and included 14 days, 1, 5, 20, 40 and 80 years.

Additionally, to characterize the plutonium density distribution across a single fuel pin in SFL 1, which was deemed important for x-ray fluorescence (XRF), each fuel pin in the infinitely reflected assembly was modeled with four independent BU rings. Given the need to quantify the consequent of shuffling an assembly throughout the core on all isotopes that impact NDA measurements, each fuel pin was modeled as one single BU region. This loss in spatial fidelity within each pin was necessary to accommodate the memory requirements for the fuel shuffling sequences, which require a greater number of burn materials.

2.1. SFL 2a, b, c Fuel Shuffling Sequences

Figure 1 shows the fuel shuffling sequence used to move fuel bundle #2 within the core, which corresponds to the fuel bundle used for isotopic information in this study. For fuel bundle #2, each pin was modeled as an independent fuel region, which due to computational limitations, still takes advantage of symmetry such that half of the 17x17 fuel assembly is modeled independently, pin by pin. Assembly 2 is located at the "Fresh UO_2 Fuel" location pertaining, and rotates through the once and twice irradiated positions. For the remaining assemblies in the core (1, 3-10) each assembly is depleted as one single burn region, where each pin is modeled separately for transport purposes.

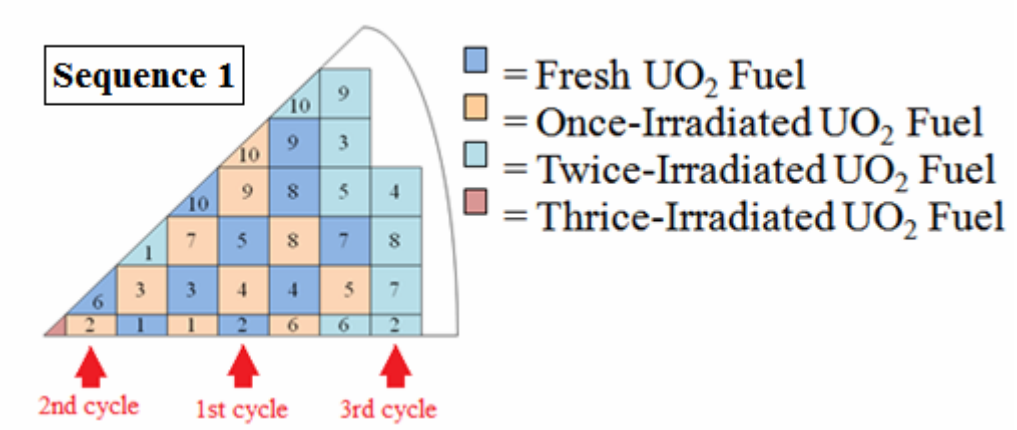

Figure 1 Fuel Shuffling Sequence 1 – Fuel Bundle #2

In addition to the initial shuffling sequence 1 shown in Figure 1, two additional shuffling sequences, 2 and 3, were simulated to gather a better understanding of what differences may arise in plutonium concentration as well as fission product distributions as a result of variations in core loading patterns; they will be included in the distribution of SFL 2b and 2c. In Figure 2, these two alternate fuel shuffling sequences are depicted and referred to as sequence 2 and sequence 3. These simulations are performed in the same fashion as the first sequence, with fuel type two having each pin depleted individually, and homogenous depletion for all other bundles, again where each pin is treated separately for transport considerations. These alternate fuel shuffling schemes were only performed for the 4% IE case, 15, 30, and 45 GWd/MTU as well as the same CTs listed above, since these were sensitivity studies intended to investigate spatial isotopic variations as a function of core loading patterns.

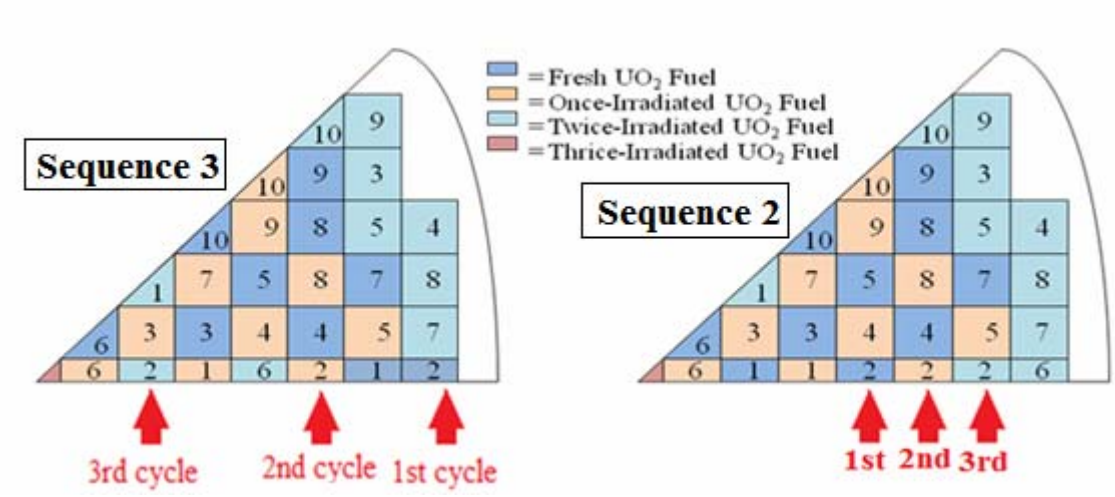

Figure 2 Shuffling Sequence 3 and 2 – Fuel Bundle #2

In traditional core loading patterns a choice of loading fresh fuel on the core periphery such as simulated in shuffling sequence 3 is a very atypical approach, however this simulation helps create a strong BU gradient across the bundle in cycle one. Thus, rotating this fuel into more reactive parts of the core serves to help quantify how strong an effect varying neutron flux gradients will have on an assembly, and in particular how strong the effect is on plutonium accumulation and the associated spatial distribution. The shuffling sequence in shuffling sequence 3, serves as a bounding condition up to 15 GWd/MTU, due to a fresh assembly adjacent to core periphery causing a high flux on the left boundary and high leakage causing a low flux off the assembly adjacent to the exterior of the core. Beyond 15 GWd/MTU for shuffling sequence 3 and for the full burn of shuffling sequence 2, a better understanding of potential variations within the domain of viable core shuffling sequences is investigated.

3. Plutonium Distribution

The plutonium mass is the quantity upon which the accountancy system in the safeguards field is based. Using the three shuffling sequences described in the preceding section the radial plutonium distribution at the end of the first cycle, as well as at the end of cycle 3, is plotted in Figure 3, Figure 4, and Figure 5, pertaining respectively to fuel shuffling sequence one, shuffling sequence three and shuffling sequence 2.

In order to display the spatial plutonium distribution across the assembly, the zero plutonium concentrations in assembly locations that hold water rods (25 locations in total) were replaced by an average of the four surrounding fuel pins so that major discontinuities did not skew the visual depiction of the elemental spatial distribution.

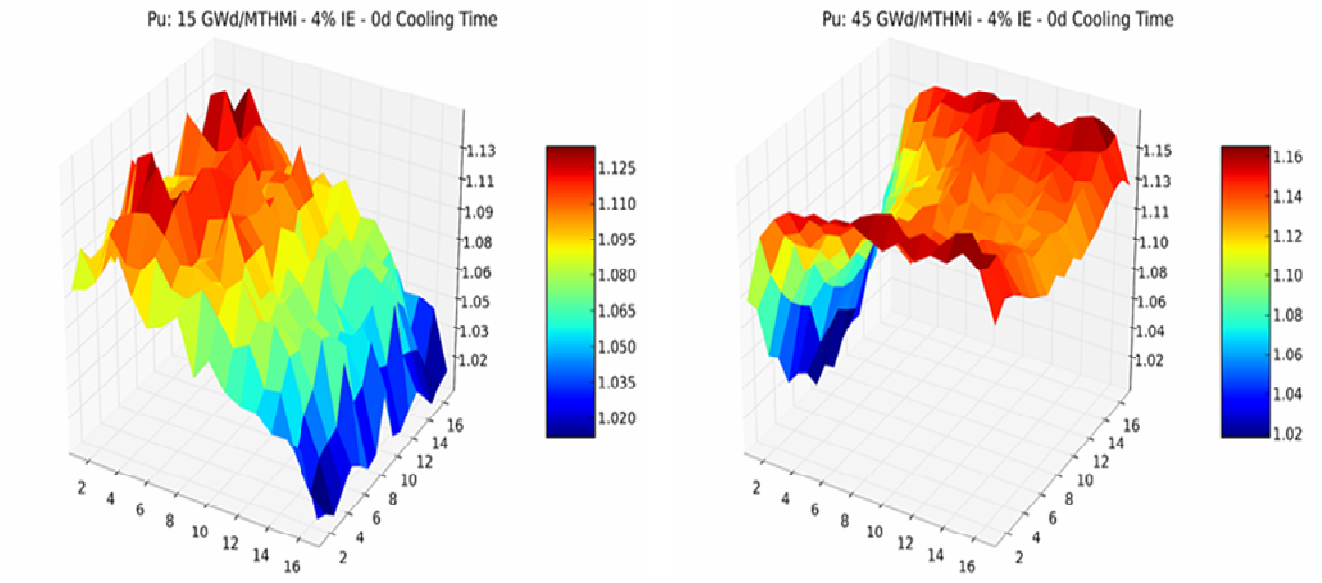

Figure 3 Shuffling Sequence 1 Pu Distribution – 15, 45 GWd/MTU

The scale on the z-axis, corresponding to plutonium mass, represents the maximum to minimum swing in plutonium mass across each assembly at each BU. This representation allows for a clear display of the magnitude of the plutonium gradient across the assembly due to the different shuffling schemes. In comparing Figure 3 and Figure 5, it is noted that in both shuffling sequences the fresh fuel started in nearly the same environment which resulted in very similar distributions at 15 GWd/MTU. In contrast, at 45 GWd/MTU the elemental gradients are somewhat mirror images. What is most interesting is that in both cases, while the spatial distribution deviated drastically being quite similar at 15 GWd/MTU and becoming close to mirror images at 45 GWd/MTU, the maximum to minimum swing in both cases was quite similar. In addition the comparison of total assembly plutonium shows a weak dependence upon the shuffling sequence, seen in Table 1 with the relative difference between the two total plutonium values being ~1% difference at both 15 and 45 GWd/MTU.

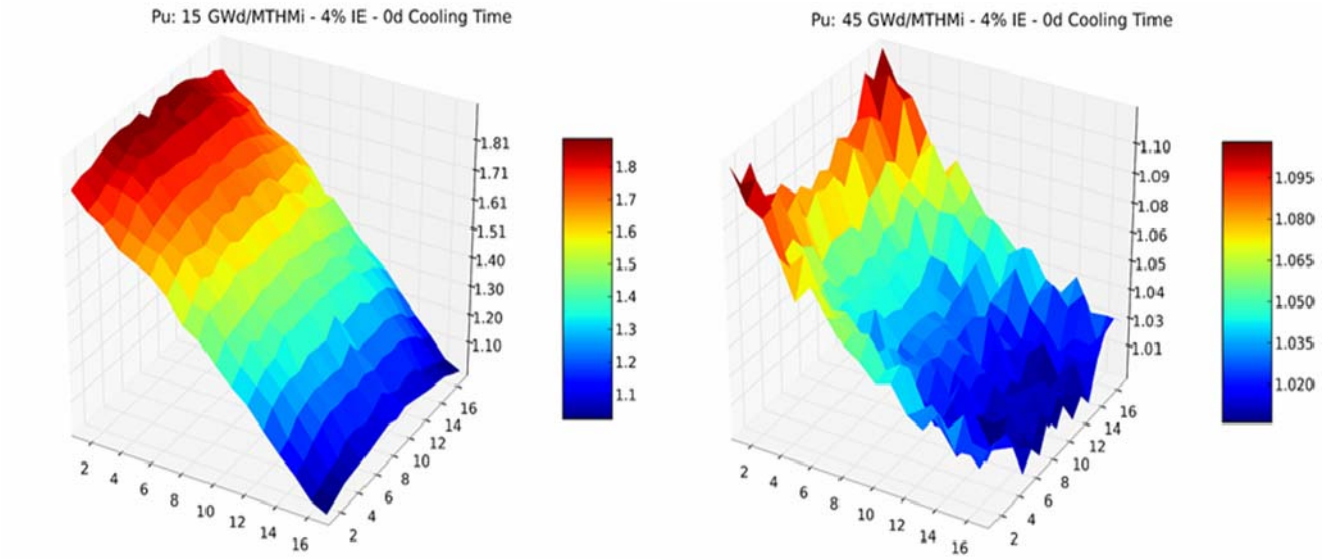

Figure 4 Shuffling Sequence 3 Pu Distribution – 15, 45 GWd/MTU

Figure 4 pertains to shuffling sequence 3 which started as fresh fuel on the exterior of the core and burned the fuel to 15 GWd/MTU before shuffling to a more internal location. While certainly a poor choice from a fuel cycle optimization stand point, this case allows for a better understanding of the effects of strong neutron flux gradients upon the accumulation of plutonium. Clearly the fuel shuffling sequence can have a significant impact upon the spatial distribution; in this extreme case at 15

GWd/MTU the plutonium swing reached ~1.8 whereas it had been ~1.13 for the two more traditional cases. This strong gradient also caused a 7.7% difference in total elemental plutonium mass compared to the total mas accumulated during shuffling sequence 1 for the same average burnup. In comparing the 45 GWd/MTU data for shuffling sequence 3 on Figure 4 and in Table 1, it should be recalled that this bundle was rotated into more central regions of the core for cycles two and three. By the time 45 GWd/MTU is reached, the maximum to minimum that had been ~1.8 had shrunk to 1.1, much more in-line with the two other shuffling sequence data at this same average BU. In addition while the relative difference between shuffling sequence 1 and shuffling sequence 3 was 7.7% at 15 GWd/MTU this value has also decreased fairly drastically to ~1.7% at 45 GWd/MTU.

Elemental Pu Mass (g)	**Shuffle 1**	**Shuffle 3**	**Shuffle 2**
15 GWd/MTU	2708.07	2499.41	2662.54
% difference		-7.70%	-1.68%
45 GWd/MTU	5024.82	4983.85	5081.89
% difference		-0.82%	1.14%

Table 1 Total Plutonium Mass (g)

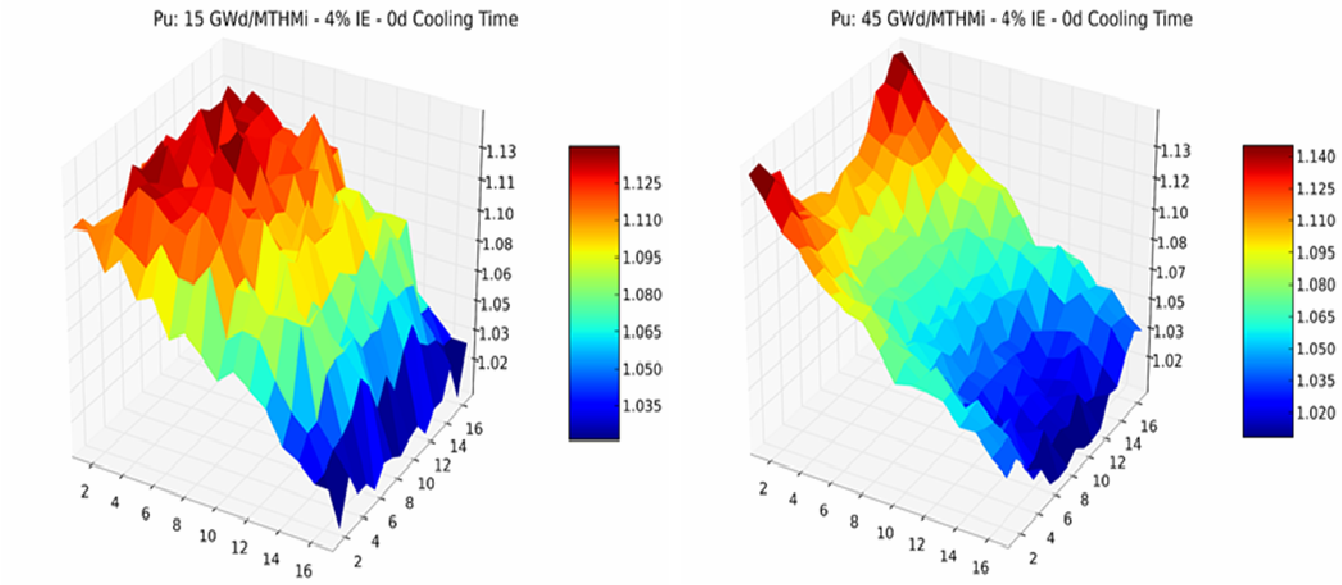

Figure 5 Shuffling Sequence 2 Pu Distribution – 15, 45 GWd/MTU

One trend consistent to all three shuffling sequences is that the highest plutonium content generally migrates to the perimeter of the assembly with increasing BU. Following the trend of the spatial distribution across the assembly, for any given row the largest elemental plutonium mass occurs at the assembly perimeter. The increased elemental plutonium mass is due primarily to the ^{239}Pu contribution, which is the single largest isotope contributing to elemental plutonium mass. The left most image in Figure 6, is for shuffling sequence 1 while the right figure pertains to shuffling sequence 3, both cases are for 45 GWd/MTU and representing the spatial ^{239}Pu distribution. The ^{239}Pu is clearly concentrated higher on the edges than the internal regions of the fuel assembly, trending with the spatial distribution across the assembly.

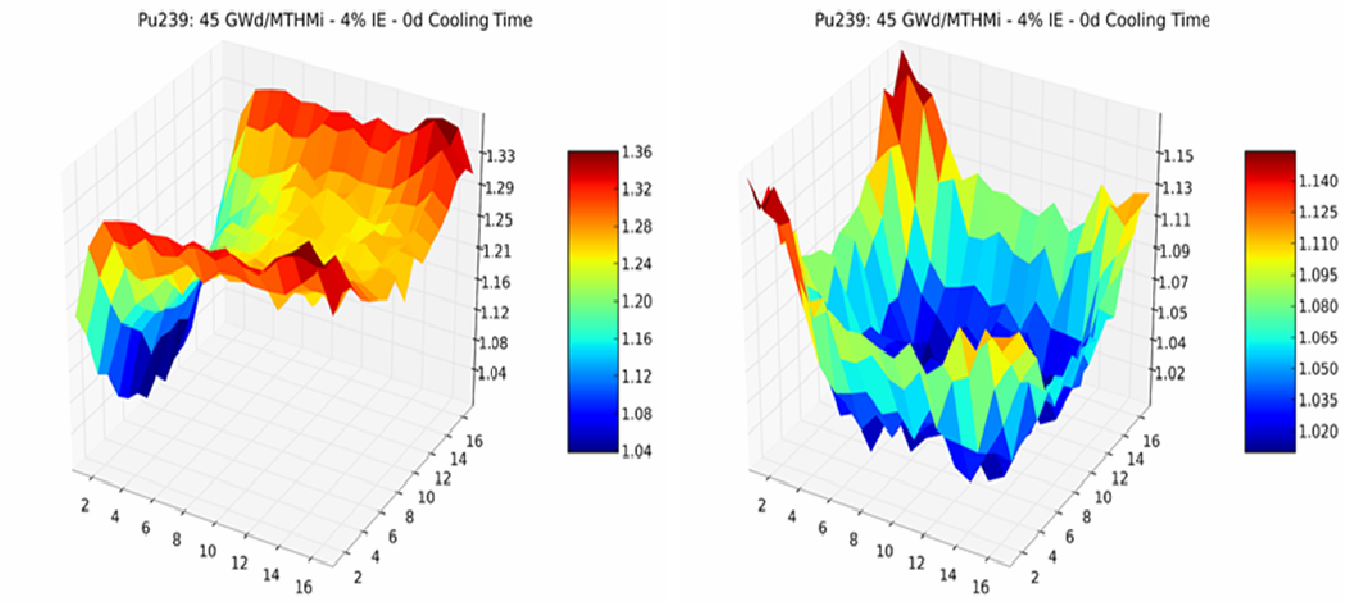

Figure 6 ^{239}Pu Concentration – Shuffling Sequence 1 (left), and Shuffling Sequence 3 (right)

Both Images at 45 GWd/MTU

This ^{239}Pu spatial distribution drives the majority of the elemental plutonium spatial distribution, thus since the ^{239}Pu is preferentially weighted towards the edge of the assembly, the same trend holds true for elemental plutonium distribution. Table 2 shows the percentage contribution of elemental plutonium that ^{239}Pu is responsible for. For all three fuel shuffling schemes at end of life, 45 GWd/MTU, ^{239}Pu accounts for slightly greater than 50% of the elemental plutonium present in this assembly. Since this ^{239}Pu contribution is quite significant, and as seen in Figure 6, this ^{239}Pu is concentrated heavily around the assembly periphery, this combination causes the trend for elemental plutonium to also be weighted towards assembly periphery. It is however noted that the peak to minimum ratio in both cases, while weighted towards the assembly periphery, still has a noticeable dependence upon fuel rotation schemes. This is seen in the maximum to minimum ratio for the two cases shown in Figure 6, with the first swing being a factor of 1.36, while the second swing a much more moderate 1.15 for 45 GWd/MTU.

45 GWd/MTU	**Sequence 1**	**Sequence 2**	**Sequence 3**
Pu (g)	5024.82	5081.89	4983.85
Pu239 (g)	2575.09	2682.90	2663.92
Pu239 % Contribution	***51.25%***	***52.79%***	***53.45%***
% Pu Change from Sequence 1		1.14%	-1.93%
% Pu239 Change from Sequence 1		4.19%	-0.71%

Table 2 ^{239}Pu - Elemental Pu and ^{239}Pu Comparison

Lastly, seen in Table 2 is the variation in total assembly plutonium mass and ^{239}Pu mass due to the different core shuffling sequences, at 45 GWd/MTU. While ^{239}Pu has a greater difference, being as much as 4.2% for sequence 2, the elemental plutonium difference was less, where the 4.2% difference in ^{239}Pu only amounted to 1.14% difference in elemental plutonium. In an opposite trend for sequence 3, the ^{239}Pu difference was less than the elemental plutonium, being -0.71% and -1.93% respectively. These results indicate that the spectral history in which the BU was accumulated has an impact in how the isotopic vectors that constitute elemental plutonium are accumulated, as well as an impact in the total mass of elemental plutonium for a given BU.

4. Passive Gamma Simulations

The use of passive gamma techniques as an NDA technique for spent nuclear fuel has been investigated and used for BU determination for several decades now[7-13]. Given this pedigree, the passive gamma approach may be useful to the NGSI effort as part of an integrated instrument intended to detect the diversion of fuel pins, while also quantifying isotopic composition and being primarily interested in elemental plutonium concentration. In support of this effort, the capability to accurately measure the BU, IE and CT of an assembly is desired. The intent is to couple this passive gamma information with simulative data from other NDA techniques to provide an accurate estimate of plutonium mass in the assembly of interest. Initial studies in support of the NGSI initiative were performed by Fensin[9][12][13] which quantified the BU and IE determination capability for the first spent fuel library which was created for an infinitely reflected 1/8 symmetric assembly. Incurring the same spatial and isotopic characteristics discussed in section two above, an amended passive gamma simulative approach needed to be applied to the assembly used in the second SFL.

Due to the asymmetric effects introduced in a fuel shuffling sequence, passive gamma simulations were required on three sides of an assembly. For an implemented system, scans might be performed either on all four sides, the four corners of the assembly, or even more locations depending on how accurate a result was needed. In SFLs 2a,b,c there was still a computational limitation on the number of burn materials allowed, thus one-half assembly reflection required simulations on only three sides of the assembly. Figure 7 shows the geometry of the simulation setup that is often used in the field. An alternative approach being considered involves a wide collimator that allows the entire side of the SF assemblies to be measured at one time, which was simulated in this study.

The passive gamma geometry setup is a difficult radiation transport problem since the photons must reach a tiny detector located a very large number of mean free paths away. Also the diameter of the collimator tube is 5.08 cm which, in the context of the size of the bundle is quite small. As Fensin[9][12][13] discussed previously, the simulative approach adopted was to tally the flux crossing the entire assembly boundary adjacent to the collimator tube. This flux was then “pushed”, or translated, up the collimator tube to the HPGE detector and a pulse-height tally was used to simulate the spectra. The same approach has been adopted for the first round of passive gamma calculations using the spatial isotopic distribution obtained from SFLs 2a,b,c.

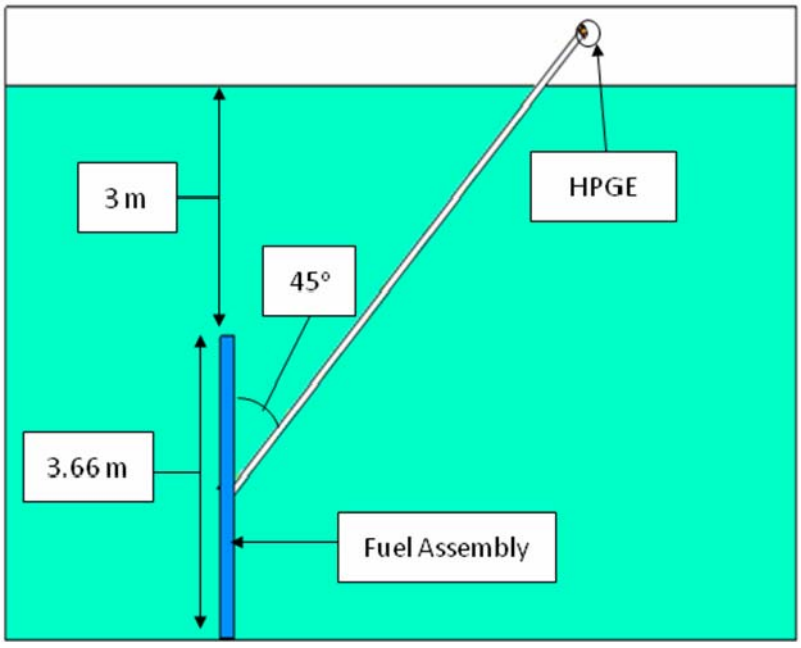

Figure 7 Passive gamma geometry

Given the simplifying assumptions made, it is expected that some inaccuracies have been introduced particularly in the magnitude of the continuum. However, given the extreme length of the collimator, the inaccuracies are not expected to be great. The resultant signal can be interpreted as proportional to the expected signal should a passive gamma scan be performed that spans the full length of each side of the assembly. In addition, a better understanding of the detection sensitivity to asymmetric effects introduced by the fuel rotation scheme, which was the initial primary objective, should result.

To tally the outgoing fluxes across assembly boundaries, a script was generated that serves as an automated process for first, computing the pin-wise gamma source file based upon the pin-by-pin isotopic compositions resulting from SFLs 2a,b,c and then creating the MCNPX input file with the combination of material compositions from SFLs 2a,b,c and the source calculation. This method is a modification and enhancement of the BAMF tool developed by Sandoval and Fensin[14]. Using this representative photon source, MCNPX tallies the energy dependent gamma lines crossing each boundary, which are then ultimately used in the creation of a final MCNPX deck which includes an f8 pulse-height tally to simulate a detectors response to the incoming gamma flux. This process can be run for every BU, IE and CT available in SFLs 2a,b,c, allowing for a wide suite of fuel rotation conditions which can serve well for assessing the potential for BU and IE determination.

Preliminary simulations have been performed to observe the variation in intensity of the gamma signal as a result of spatial BU distributions. The first case chosen to simulate was the 15 GWd/MTU case from shuffling sequence 3, data which was already illustrated in Figure 2 and Figure 4 above. This was chosen since it has the most extreme spatial gradient of any of the shuffling rotations simulated. Figure 8 shows the relative intensity spectrum for the five year CT case at 15 GWd/MTU. Although it is difficult to see due to the logarithmic scale, the detector associated with “side 2”, corresponding to the highest burnt side of the assembly, also experienced the strongest gross signal.

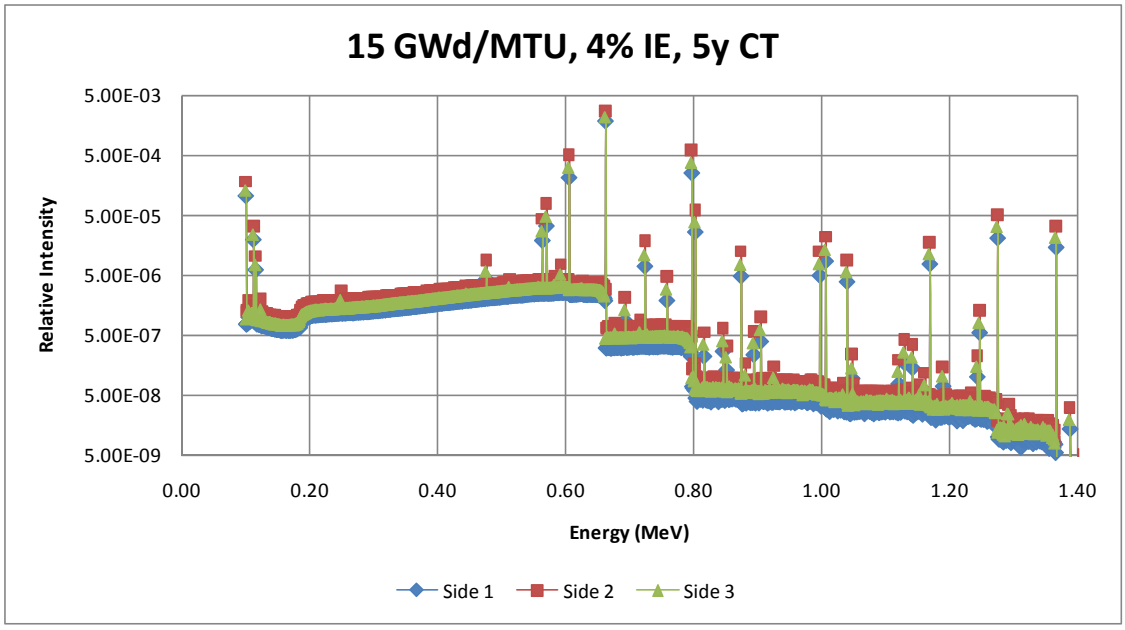

Figure 8 Relative intensity vs. energy (MeV) – 15 GWd/MTU – 5y CT

Relative difference from side 2 activity (5y cooling time)		
	Side 1	Side 3
Cs-134 (.6047 MeV)	-40.86%	-58.84%
Cs-137 (0.6617 MeV)	-23.86%	-32.49%
Cs-134 (0.7959 MeV)	-39.55%	-58.39%

Table 3 Relative difference – 15 GWd/MTU – 5y CT

In quantifiable terms, Table 3 shows the percentage differences for the three most prominent peaks, the 662 keV line from ^{137}Cs, and two ^{134}Cs lines at 605 keV and 796 keV. From Table 3 it is apparent that the ^{134}Cs peaks have a stronger dependence on the assembly spatial power distribution than the ^{137}Cs peak, having nearly double the percentage difference than the variation seen in the ^{137}Cs peak intensity. This result is as expected since ^{134}Cs accumulation scales closely with the square of the flux, whereas the ^{137}Cs accumulation scales linearly with the flux.

Equation 1 and Equation 2 show the decay chains upon which ^{137}Cs and ^{134}Cs production depend, keeping in mind that both chains are initially dependent upon the flux through the fission product yields of each isotope in the decay chains. In the case of ^{134}Cs the second dependence on flux comes through the capture reaction of ^{133}Cs to ^{134}Cs, and this additional dependence causes the greater variation in signal intensity for ^{134}Cs seen in Table 3. The edge of the bundle that was located on the core periphery received much less of a flux intensity than the internal edge due to leakage, causing the greater signal intensity differences in ^{134}Cs when compared to ^{137}Cs which only depends on the flux through fission yields.

$$^{137}_{I} \rightarrow {}^{137}_{Xe} \rightarrow {}^{137}_{Cs} \rightarrow {}^{137}_{Ba^{m}} \rightarrow {}^{137}_{Ba}$$

Equation 1 ^{137}Cs production chain

$$^{133}_{I} \rightarrow {}^{133}_{Xe} \rightarrow {}^{133}_{Cs}(n,\gamma)^{134}_{Cs} \rightarrow {}^{134}_{Ba}$$

Equation 2 ^{134}Cs production

Figure 9 shows the relative gamma intensity as a function of energy for the 45 GWd/MTU, 4% IE and five year CT case from the shuffling sequence 1. Quite different from the preceding spectrum, the minimum-to-maximum swing in plutonium concentration for this case, seen in Figure 3, was much lower than the preceding case, being on the order of 15% as opposed to 80%. This indicates a more even power distribution across the assembly, which is also observed in Figure 9. Here it becomes very hard to visually distinguish the three spectra from each other, where the side 1 spectrum is, for the most part, hidden behind the other two.

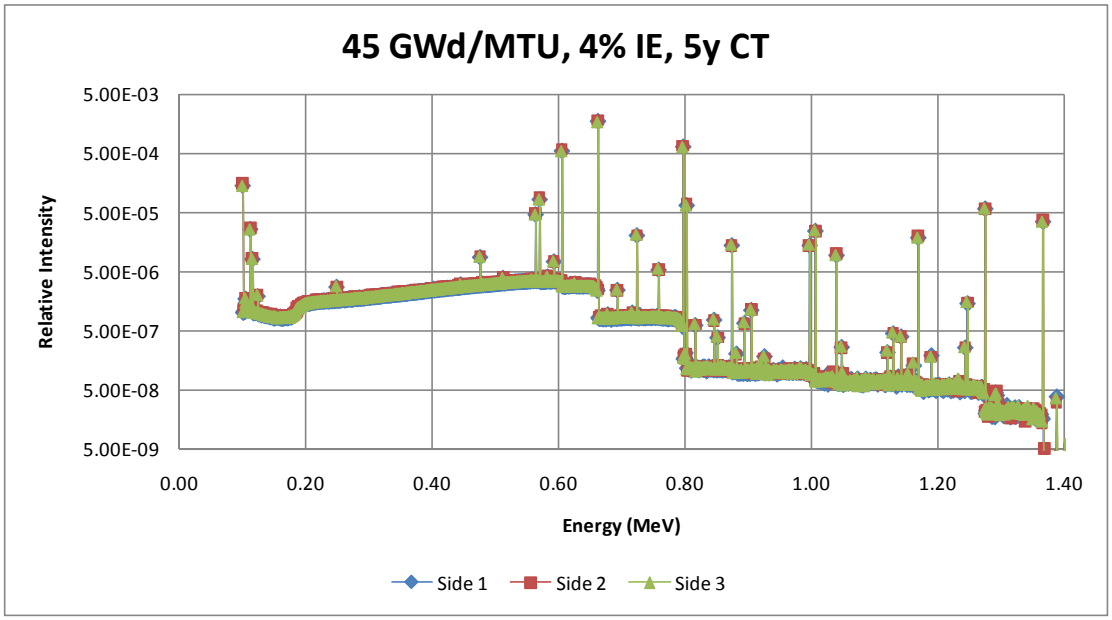

Figure 9 Relative intensity vs. energy (MeV) – 45 GWd/MTU – 1y CT

The percentage differences, relative to the maximum activity which again occurred across side 2, are much lower than in the previous case. Observed in Table 4, for all three isotope lines the largest difference occurred on side 1, varying from ~5-7%. There is clearly spatial sensitivity, largely influenced by the core loading patterns, which was also observed by trending the differences at each BU step for shuffling sequence 1. In this case differences of ~16% were observed for ^{134}Cs lines, and 8% for ^{137}Cs at 15 GWd/MTU. After the fuel was shuffled and continued burn to 30 GWd/MTU, the bundle moved to a further centrally located slot which served to create a more evenly distributed burn, and differences in ^{134}Cs lines have been reduced to ~4% with ~2.5% differences observed in ^{137}Cs. From there the bundle was shuffled to the core periphery where the differences grew, due to the exterior of the bundle having a large leakage term with virtually no incoming flux. The percent differences observed here were 5-7% in both ^{134}Cs and ^{137}Cs.

Relative difference from side 2 activity (5y cooling time)		
	Side 1	Side 3
Cs-134 (.6047 MeV)	-6.51%	-3.88%
Cs-137 (0.6617 MeV)	-5.21%	-4.51%
Cs-134 (0.7959 MeV)	-5.86%	-3.40%

Table 4 Relative difference – 45 GWd/MTU – 5y CT

With a half-life of 2 years, a significant portion of the ^{134}Cs has decayed by the time the 5y data was extracted, thus intensities are greater for gamma lines from ^{134}Cs at shorter CTs. Also, while there is a clear dependence upon the core shuffling sequence such that signal intensity differences amongst the sides of the bundle may be moderately larger at a higher BU compared to a lower BU, there also is a general trend of the relative differences amongst the sides trending from greater values to lesser values as BU increases, which arises from the reactivity characteristics of a bundle in the core. If one part of the bundle has been burned at a faster rate, in general as the bundle rotates throughout a typical shuffling sequence, the BU distribution across the bundle will tend to smooth out since there will be more fissile material in the under burned part of the bundle and power generation will eventually shift to the under-burned portion of the bundle. Again this is somewhat dependent upon shuffling sequence, but will hold true for typical fuel shuffling schemes. Thus, typically, high BU assemblies will not have the largest differences in signal intensity across the bundle from passive gamma measurements unless anomalous fuel shuffling practices are employed. This effect is observed in the case of the three BU points for shuffling sequence 1. The bundle rotated from one central location, to a more central location, til finishing its life on the core periphery. Despite the fact that strong asymmetries were experienced in the final shuffling sequence where high leakage occurred on one boundary, the relative difference at 45 GWd/MTU was significantly less than at 15 GWd/MTU, being ~16% at 15 GWd/MTU compared to ~6% at 45 GWd/MTU.

Comparing the ratio of ^{134}Cs/^{137}Cs a clear spatial dependence is evident for the strongly asymmetrically burned assembly. Using shuffling sequence 3 results at 15 GWd/MTU shuffling sequence 1 results at 45 GWd/MTU, Table 5 shows the calculated Cesium ratios for these two cases. The data used to compute the ^{134}Cs contribution was a sum of all gamma lines emitted. The ratios from shuffling sequence 3 have a large variation, with side 1 being nearly 40% less intense than side 2, and side 3 being ~20% less intense than side 2. In contrast, shuffling sequence 1 only had a maximum difference of 2% between the most intense and least intense signals. Using this data combined with the isotopic information of each pin, it is possible that a relationship between the Cesium ratios and the plutonium content of the pins contributing to the signal could exist, allowing the estimation of plutonium content in the fuel pins contributing to the passive gamma signal.

Sequence 3 - 15 GWd/MTU - 5y			
	cs134	cs137	**ratio**
Side 1	5.43E-04	1.84E-03	**0.2955**

Sequence 1 - 45 GWd/MTU - 5y			
	cs134	cs137	**ratio**
Side 1	1.37E-03	1.69E-03	**0.8100**

Side 2	1.31E-03	2.72E-03	**0.4801**
Side 3	7.84E-04	2.07E-03	**0.3784**

Side 2	1.46E-03	1.79E-03	**0.8178**
Side 3	1.41E-03	1.71E-03	**0.8258**

Table 5 Cesium Ratios

5. Future Work

Much work has gone into the generation of SFLs 2a,b,c, in an attempt to generate source signals that closely represent true conditions expected from a PWR 17x17 assembly. In leveraging this library for plutonium distribution studies it was observed that as BU increases, plutonium content not only increases but tends to have higher concentrations on bundle edges, and particularly bundle corners. Using known plutonium concentration from SFL 1 and SFLs 2a,b,c, particularly the edge concentration, to predict internal plutonium concentration, both in a spatial pin-by-pin distribution as well as a bundle total quantity of plutonium from the edge plutonium concentration would be useful in support of additional integrated instrument design in support of the NGSI initiative, particularly related to XRF instrument design and assessment. In addition, while passive gamma simulations of the total edge gamma flux benefit in understanding source magnitude differences as a result of shuffling schemes, more concentrated simulations attempting to simulate what the signal the HPGE detector would see would be beneficial, and allow the better estimations of how many locations, and which locations, would be needed to reliably extrapolate from passive gamma signal to an estimation of assembly average BU. Clearly multi-sided simulations would need to be performed for this task, but how many locations per side are needed, and where are the most important points to scan? These questions would need to be addressed to attempt to reliably use this technique to predict assembly average BU. In addition there is also a potential for using the passive gamma signal to calculate Cesium ratios which may be provide an accurate estimate for plutonium concentration in the fuel pins contributing to the passive gamma signal. A characterization of which pins contribute, and how much each pin contributes, to the total passive gamma signal for a given geometrical set up needs to be performed to better estimate the total plutonium content in those contributing pins.

6. Acknowledgements

The authors would like to acknowledge all those who have contributed to the NGSI effort, in particular to the Next Generation Safeguards Initiative, Office of Nuclear Safeguards & Security (NA-241), National Nuclear Security Administration in the U.S. Department of Energy for their support throughout this effort.

7 Legal matters

7.1. Privacy regulations and protection of personal data

"I agree that ESARDA may print my name and contact information as well as this paper in the ESARDA Bulletin and/or Symposium proceedings or any other ESARDA publication, and when necessary for any other purposes connected with ESARDA activities."

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Detection of Partial Defects in LWR Spent Fuel Using a DCVD

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Abstract:

The inventory of stored spent fuel assemblies is reaching the capacity of many fuel ponds. To alleviate this congestion, spent fuel assemblies will need to be transferred to dry storage containers. Prior to this operation, there is a safeguards requirement to verify that the spent fuel is intact and that no fuel rods have been diverted for clandestine activities. This paper reports on recent research on the ability of the Digital Cerenkov Viewing Device to detect the diversion of fuel rods. Modelling has been carried out for a number of diversion scenarios to determine the impact on Cerenkov light intensity. Field studies have also been carried out on Boiling and Pressurized Water Reactor fuel to determine whether it is possible to detect changes in light intensity as a result of fuel rod diversion. The results of these studies are given in this report.

Keywords: DCVD; Cerenkov; partial defects

1. Introduction

The measured light intensity in the DCVD Cerenkov image (Figure 1) of spent fuel is quantitative. Each image pixel contains an intensity value that is proportional to the number of Cerenkov photons generated by a spent fuel assembly. Each Cherenkov photon is an indirect measure of the fission product concentration in the spent fuel assembly. Using this knowledge, we can examine a spent fuel image quantitatively to determine whether it has fuel rods missing or replaced with non-irradiated steel rods.

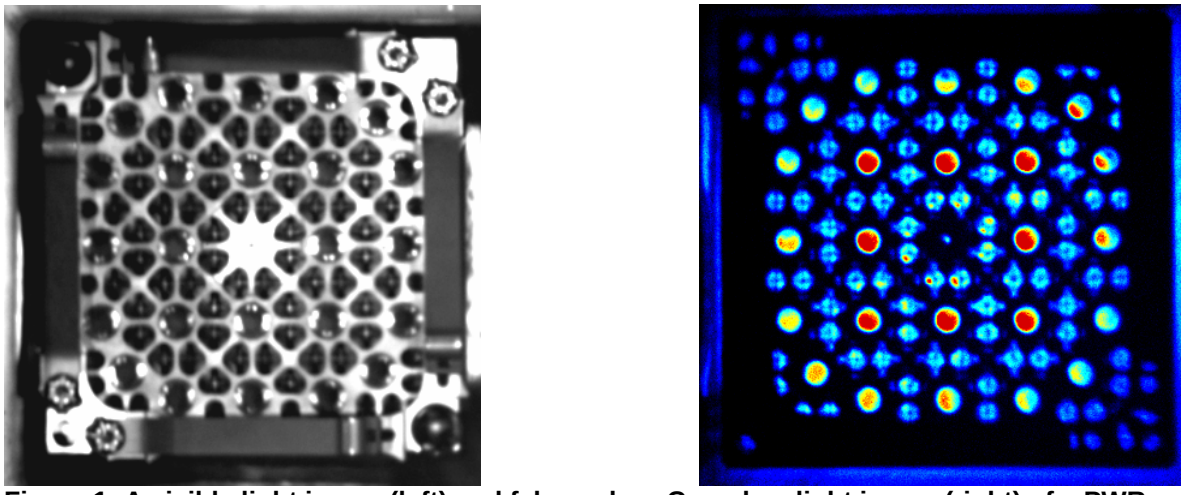

Figure 1: A visible light image (left) and false colour Cerenkov light image (right) of a PWR spent fuel assembly

Previous studies carried out[1] on fuel assemblies with missing and substituted fuel rods showed that if the location of the missing fuel rod is not obscured by the top plate, a single missing fuel rod can be

detected visually. Close visual examination of the water gaps around a single substituted fuel rod shows a decrease in light intensity[2]. Analysis of the measured light intensity in the water gaps around the substituted fuel pin shows a significant drop in light intensity when compared to the light intensity of the water gaps at similar positions in the fuel assembly.

These initial studies showed that detection of a single fuel rod anomaly was possible but not practical. Another approach was needed to detect partial defects.

2. Detecting Partial Defects Using Quantitative Methods

The current IAEA definition of a partial defect in spent fuel is 50 percent of the fuel rods have been removed or have been replaced with non-fuel rods. The case where 50 percent of the fuel rods are missing can easily be detected visually so this research focused on the more difficult case where 50 percent of the fuel rods are substituted with blackened non-fuel rods.

The number of Cerenkov photons generated by a fuel assembly is proportional to the concentration of fission products. These fission products generate gamma rays which result in Cerenkov light emission via Compton scattering. The relationship between Cerenkov photon flux and burnup and cooling time for BWR fuel is shown in Figure 2. Curves are generated for burnups ranging from 2 500 to 50 000 MWd/t U and for cooling times from one to 40 years. These results are based on theoretical calculations by S. Rolandson[3] and field measurements by L. Ilver[4]. The assumption was made that the lower concentration of fission products in a spent fuel assembly with replaced rods would generate less Cerenkov light and that the quantitative measurement of Cerenkov light by the DCVD was sensitive enough to detect this change.

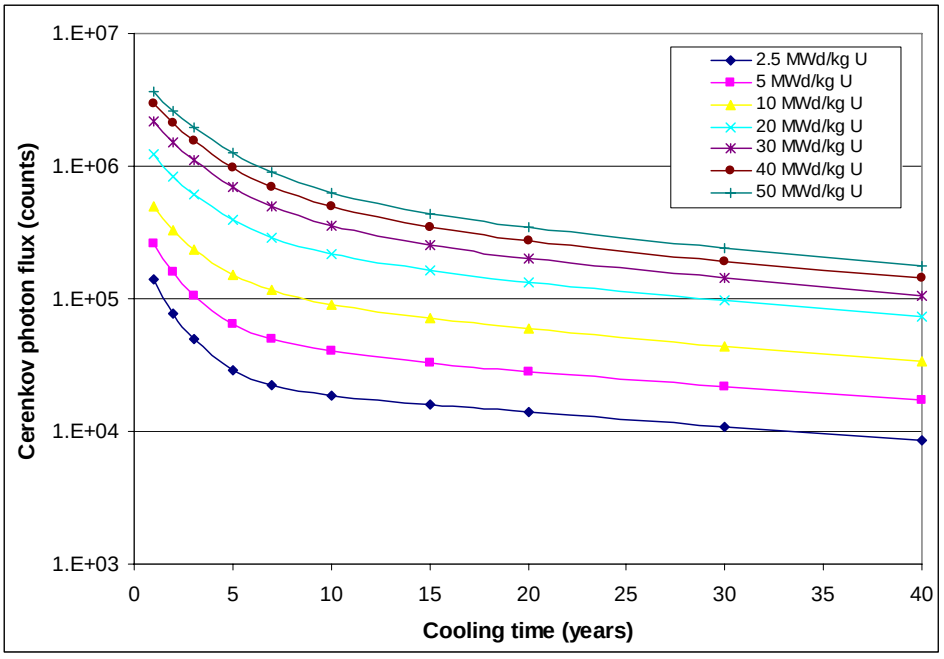

Figure 2: Cerenkov photon flux expected from a given burnup and cooling time

2.1 An algorithm for measuring Cerenkov light

The algorithm for assessing Cerenkov light intensities from spent fuel assemblies described in this report is a relative process where the consistency of measurements from a group of fuel assemblies is examined to determine if a partial defect exists. It is possible to obtain absolute intensities from spent fuel but this requires calibration of the DCVD and significant effort to account for water quality, ambient lighting and measurement reproducibility. The relative method eliminates these common factors between fuel assemblies.

The Cerenkov light intensity of a DCVD measurement of a fuel assembly is calculated using the following procedure:

- A region of interest (ROI) is selected which includes the fuel assembly and as little of the surrounding area as possible (Figure 3).
- The background level within the ROI is obtained by finding the minimum average intensity of each 3x3 pixel area in the selected ROI. This background value consists of the detector base level signal, read-out noise, stray light from ambient lighting and scattered Cerenkov light from fuel assemblies in the water pond. The background intensity value is subtracted from each pixel in the ROI to obtain a background corrected intensity value.
- The corrected intensity value from all the pixels in the ROI are then summed to produce a Cerenkov light intensity value for the measured fuel assembly.

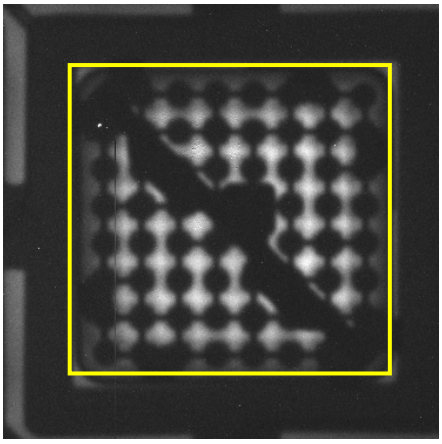

Figure 3: The Region of Interest (ROI) surrounding a BWR spent fuel assembly

The DCVD measurements are processed to obtain the Cerenkov intensity from each fuel assembly. The expected intensity for each fuel assembly is calculated from the theoretical curves shown in Figure 2. The two sets of data are entered into a spreadsheet and a scaling factor is obtained to normalize the measured data to the expected intensities.

The difference between the measured intensity and expected intensity should be less than the minimum deviation caused by a 50 percent partial defect. The deviation is derived via modelling as there are no available assemblies with a 50 percent partial defect to reference.

2.2 Modelling partial defect effects in a fuel assembly

The modelling efforts focused on a 30 percent partial defect because it was felt that this level of detection could be achieved. Fuel rod substitution with black stainless steel rods at the 30 percent level in spent fuel was expected to produce a 30 percent reduction in Cerenkov light intensity. Modelling studies were carried out on a PWR 17x17 fuel assembly and a BWR 8×8 fuel assembly to determine the Cerenkov light characteristics. The PWR study is discussed in detail. Since the BWR modelling methodology is similar, only the results are presented.

A Framatome 17×17 AFA-3GA fuel assembly with a burnup of 44 931 MWd/t U, and cooled for 2.3 years was modelled. This AFA-3GA fuel assembly contains 264 fuel rods and 25 guide tubes and was selected because it had been studied with the DCVD and a comparison of the model with a real assembly could be made. The fuel depletion code ORIGEN-ARP was used to calculate the gamma energy spectrum of the fuel assembly. Geant 4 was used to calculate the Cerenkov light production and Zemax was used to trace the Cerenkov photons from the fuel assembly to the detector. The spacer grids and top nozzle and substituted fuel pins are included in the models and all mechanical parts are assumed to be stainless steel and black and totally light absorbing.

Substitution of 30 percent of the fuel rods required replacement of 80 fuel rods in the PWR fuel assembly. To determine if all substitution scenarios result in the same decrease in Cerenkov light reaching the DCVD, three substitution scenarios were modelled:

- 80 substituted rods under the lifting handle
- 80 substituted rods interspersed in the central area of the assembly (not under the handle)

- 40 substituted rods in one quadrant but not under the handle.

In the last case, only 40 substituted rods were used because that defines the visible area of the quadrant. The PWR and BWR modelling results are shown in Table 1.

PWR modelled case 17×17 fuel	Intensity decrease	BWR modelled case 8×8 fuel	Intensity decrease
80 steel rods interspersed (30%)	40%	19 steel rods interspersed (30%)	26%
80 steel rods under the handle (30%)	15%	19 steel rods under the handle (30%)	23%
40 steel rods in one quadrant (15%)	22%	9 steel rods in one quadrant (14%)	22%

Table 1: Intensity decrease relative to reference in PWR and BWR modelling scenarios

When the 80 steel rods are placed under the handle in a PWR assembly the decrease in intensity is only about 15 percent. This difference would be difficult to detect in view of the alignment precision of ±5 percent. If the substitution is increased to 50 percent, which corresponds to 132 fuel rods, the decrease in light intensity is expected to be approximately 25 percent which makes the detection of this partial defect more likely. The decrease in light intensity for the other cases is significant and detection would be expected at the 30 percent substitution level.

The 8×8 BWR fuel was selected for modelling because this fuel design is one of the original and is the most frequently encountered design. Substituting 30 percent of the fuel rods with stainless steel rods would result in an intensity decrease that should be detected. If the modelling is extrapolated to a 50 percent substitution, a partial defect should be readily detected in all scenarios.

The most difficult case to detect with PWR fuel appears to be when the partial defect occurs around the perimeter of the assembly. The first issue is that all of the perimeter fuel rods are hidden by the top nozzle. Secondly, when the DCVD is aligned over the centre of the fuel assembly, the lower part of the perimeter rods are shielded from view by the inner rods, reducing their contribution of Cerenkov light.

Fortunately, this diversion scenario can be detected by lifting the assembly 2 to 3 meters. The substituted rods around the perimeter of the assembly will not emit Cerenkov light.

3. Field test results

The methodology described here was tested during field campaigns at Ascó, Spain on PWR fuel and at Chin Shan, Taiwan on BWR fuel.

A total of 156 PWR fuel assemblies were measured at Ascó, Spain for their Cerenkov intensities over a two day period. Fifty-six of the fuel assemblies had inserts (plugs, burnable poison and control rods). The burnup ranged from 24 000 to 55 000 MWd/t U and the cooling times ranged from 2.3 to 23.6 years.

The expected intensity value for each fuel assembly was determined from the theoretical intensity curves and a scaling factor was calculated to scale the theoretical values to the measured intensities. A lower boundary level of 30 percent decrease from the expected intensity is drawn on the plots. An upper boundary of 30 percent is also drawn on the plots to bracket the results.

Figure 4 shows the higher burnup, shorter cooling time assemblies and includes assemblies with and without inserts. The assemblies are ordered from highest to lowest expected intensity based on the curves shown in Figure 2.

The measured intensity values for assemblies without inserts are between the 30 percent boundaries indicating no partial defect. For assemblies with inserts, the measured Cerenkov intensity is about 30 percent lower because the light from the guide tubes is blocked. Only one fuel assembly without an insert is at the 30 percent boundary, AE21, which may be due to interrupted irradiation (still under investigation). This situation occurs when a fuel assembly is taken out of the core before terminal burnup and is cooled for one or more years and then put back into the reactor and irradiated to its terminal burnup. There is a group of 2.3 year cooled fuel assemblies that have much higher

intensities than expected. A possible explanation is that there are slight design changes in the fuel assembly that result in a higher than normal light output.

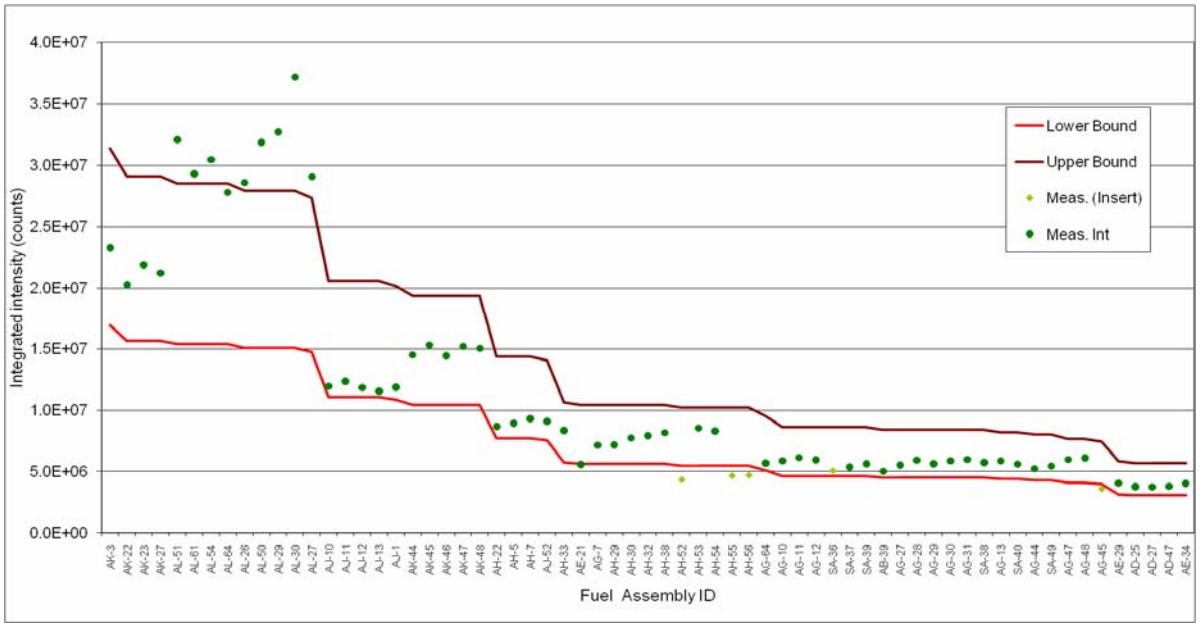

Figure 4: PWR fuel measurements with 30% lower boundary

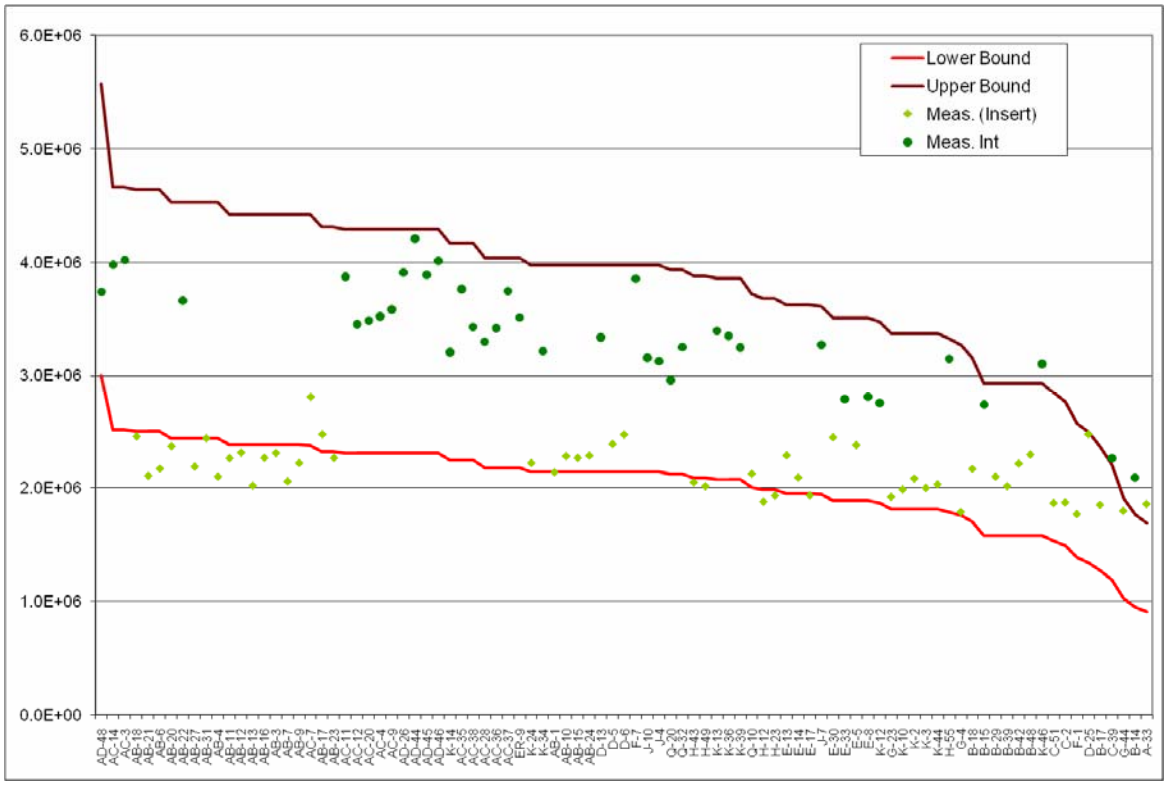

Figure 5: Long cooled PWR fuel assemblies with 30% lower boundary

Figure 5 shows the lower intensity (longer cooled) fuel also ordered from the largest expected intensity to lowest expected intensity. The data also show good agreement between the measured and calculated values. The values below the 30 percent boundary are all fuel assemblies with inserts which reduces the light intensity from the fuel assembly. There appears to be no evidence of a partial defect in these assemblies.

A total of 140 BWR fuel assemblies were measured at the Chin Shan NPP. The burnup of these assemblies ranged from 1200 to 34 700 MWd/t U and cooling times ranged from 11 to 30 years. The expected Cerenkov intensities were calculated in the same manner as in the Ascó case and the same procedure was used to produce a scaling factor to normalize the expected Cerenkov intensities. The results from the measurement of 140 fuel assemblies are shown in Figure 6 ordered from lowest expected intensity to highest.

There are only three fuel assemblies that are outside of the 30 percent limit, representing about 2 percent of the 140 fuel assemblies. The cause of these outliers has not yet been investigated

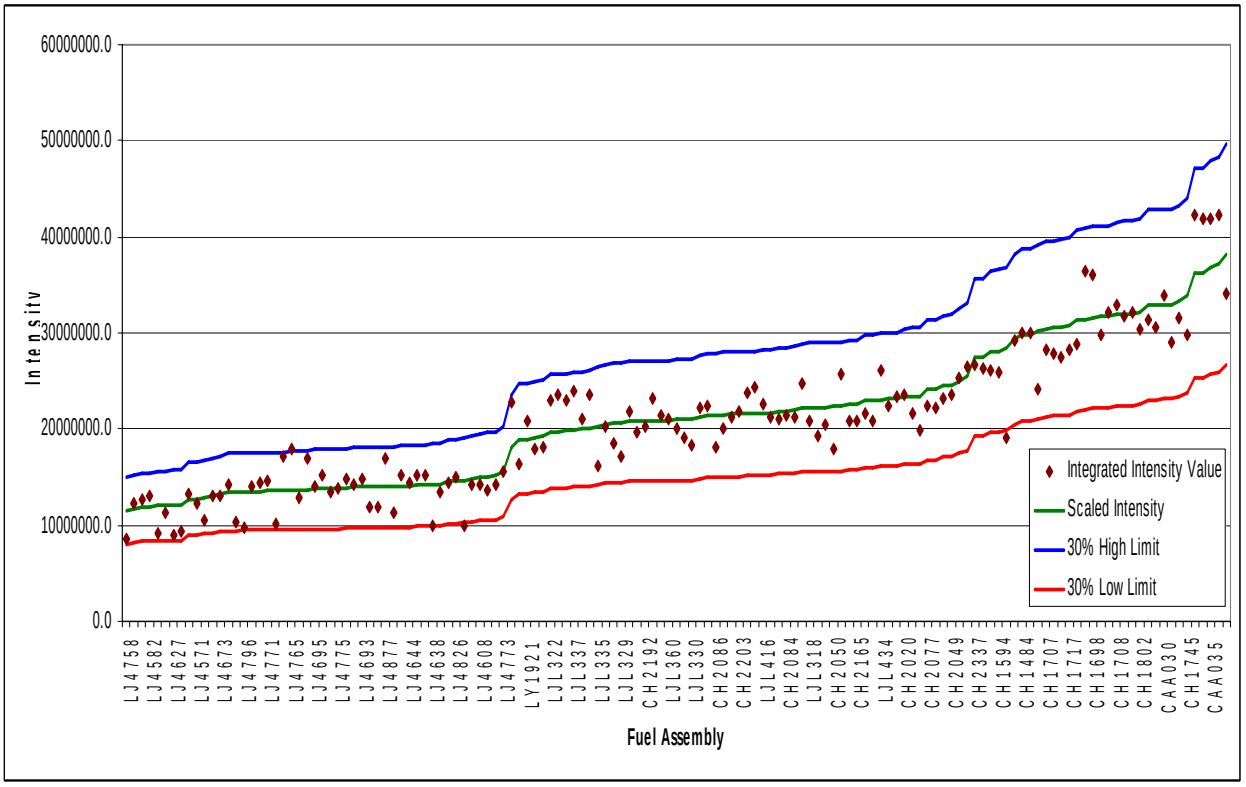

Figure 6: BWR fuel measurements with a 30% lower boundary

4. Incorporation of the methodology into the DCVD software

The apparent success of the methodology prompted the incorporation of the method into the DCView program which forms the user interface for the DCVD. DCView was modified to import a spreadsheet format file containing the operator declaration of burnup and cooling time along with the fuel identification, fuel location and expected intensity.

The user interface was modified to display this information while the inspector measures and moves between fuel assemblies. The internal code was also modified to calculate the measured intensity within the region of interest (ROI) and produce a report of the measured intensity and relative intensity. If the measured intensity falls 30 percent from the expected value, the inspector is alerted with a warning. Because this is a relative method, several assemblies must be measured before the partial defect detection algorithm can recognize an outlier.

These new software features were tested at a field trial involving IAEA, Euratom, and SKB personnel. Since a fuel assembly with a “partial defect” was not available, the defect was simulated by increasing the declared burnup of one of the assemblies. This has the effect of raising the expected intensity which was expected to generate an outlier. The participants in the exercise were not informed of the modification.

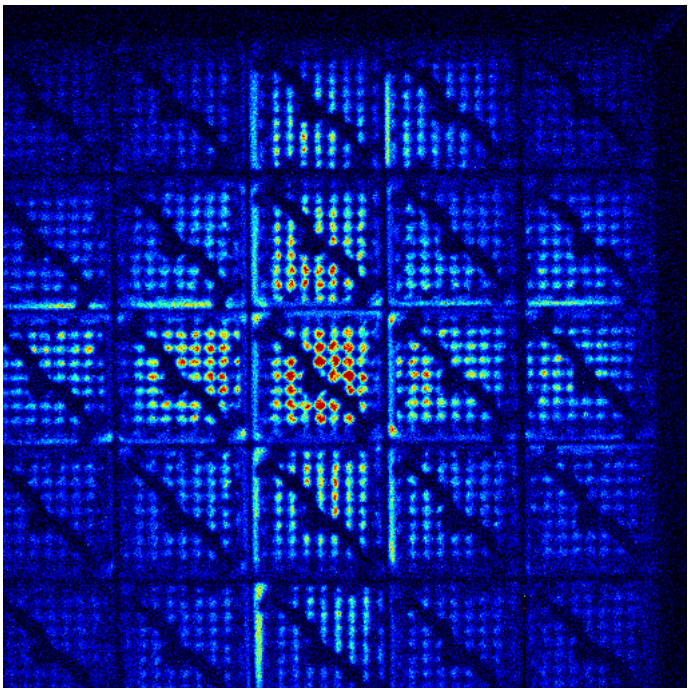

Figure 7: Cerenkov false colour image of a basket of BWR fuel

Each of the three groups of participants measured the basket of 8x8 BWR fuel (Figure 7). The deviation from the expected intensity was detected by DCView and a warning was issued that an outlier was detected and the fuel assembly information was highlighted on the user interface (Figure 8).

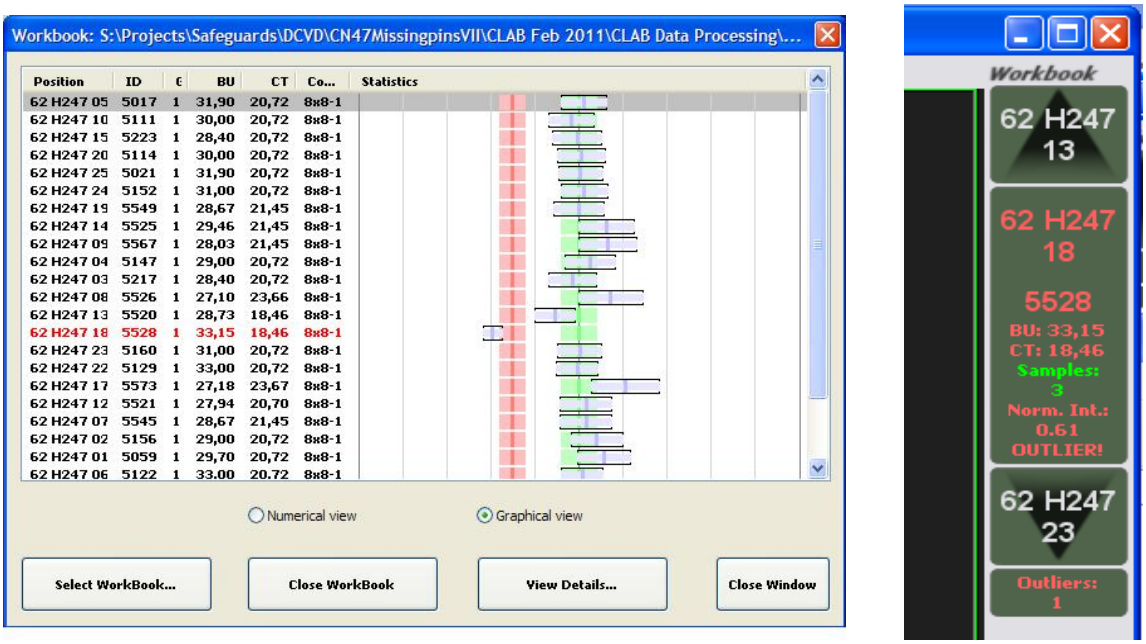

Figure 8: DCView on screen indicator of an "outlier". In the graphical view the suspect assembly is highlighted in red and on the status panel of the user interface the assembly information is highlighted in red as well

5. Conclusions

Modelling has shown that a 50 percent partial defect results in a measureable decrease in the measured Cerenkov intensity for even the most difficult scenarios. For PWR spent fuel these scenarios occur when the substitution is in the perimeter of the fuel assembly which is shielded from view by the lifting handle and top nozzle. For BWR the most difficult substitution to detect appears to be when the some of the substituted rods occur under the lifting handle with others around the perimeter.

Field studies on PWR and BWR fuel using the proposed method to detect partial defects result in a 1 to 2 percent false positive rate.

Implementation of this process into the DCVD software has been completed and field tested with good results. Further study and field testing is planned to increase the confidence in the results.

6. Acknowledgements

The authors would like to acknowledge the support of the operators of the Ascó Spain NPP, Chin Shan Taiwan NPP, and CLAB facility in Sweden for their cooperation and support in this research. We would also like to acknowledge the support of the IAEA in arranging the Chin Shan field trial and for their input and support.

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Spent Fuel Measurements with the Fork Detector at the Nuclear Power Plant of Doel

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Abstract:

SCK•CEN is involved in the investigations and implementation of experimental methods for the burnup determination of spent fuel elements. One of these methods is the so-called Fork Detector, consisting of two cadmium wrapped polyethylene arms containing one fission chamber and one ionization chamber each. In the case of a spent fuel element (SFE), the measured neutron count rate is mostly due to the neutron emission from the spontaneous fission of ^{244}Cm, taking into account a sufficient cooling time; this is related to the burnup by a power function whose parameters can be determined experimentally by measuring SFEs with well-known initial enrichments and irradiation histories. Once these parameters are known the burnup can be determined from the measured neutron count rate.

In the past this detector has been successfully used for the burnup determination in the Nuclear Power Plant (NPP) of Tihange (Belgium). This detection system was refurbished and upgraded to fit the requirements of the nuclear power plants (NPP) of Doel (Belgium). In particular efforts were devoted to develop a new mechanical tool and rack support for the installation and fixation of the Fork detector in the pond and a user-friendly software for the data acquisition and analysis.

This paper reports about the measurement and qualification campaigns that were carried out at the NPP of Doel and the results obtained, and highlights its use for satisfying the safety requirements of the Authorities and reprocessing industry, related to reprocessing of spent fuel.

Keywords: Spent Fuel; Fork detector; Burnup determination; Non Destructive Assay

1. Introduction

This paper focuses on the use of the so-called Fork detector [1,2] for spent fuel measurements aimed at the determination of burnup. The Fork detector in use was developed at SCK•CEN and installed at the Nuclear Power Plant (NPP) of Tihange (Belgium) where it was in use between 1992 and 1994. Recently SCK•CEN refurbished this detection system which was then installed in the deactivation spent fuel ponds of the NPP of Doel (Belgium). A total of four reactor units are present in the NPP. The Fork detector described in this paper is meant to be used in the deactivation spent fuel ponds for the units Doel3 and Doel4 indicated with D3 and D4 in the text.

The Fork detector is an instrument in use for the Safeguards inspections carried out by IAEA and Euratom to verify that there has been no diversion of (part of) a SFE [1]; this is done by assessing the burnup of a SFE [2]. In Belgian NPPs, the assessment of the burnup at the extremities of a SFE is of interest to satisfy safety requirements, related to the reprocessing of the spent fuel. Burnup measurements are required for a SFE with an initial enrichment of at least 4.20% to verify that the minimum burnup in the end regions of the assembly is 11 GWd/tU.

In the first part of the paper the measurements principles are described; the main features of the detection system and of the refurbishment follow; at the end of the paper, the results of calibration measurements are also given.

2. The use of the Fork detector for spent fuel measurements

A lot of effort has been devoted in the past to investigate methods to determine the burnup of spent fuel assemblies in a non-destructive way [3]. More recently SCK•CEN investigated experimental methods to assess their capability for burn up determination [4]. A significant effort on spent fuel measurement techniques is also being carried out in the U.S., under the framework of the Next Generation Safeguards Initiative [5,6] and is reported in other papers presented at this Symposium.

One of the methods applied is isotopic correlation, based on the measurement of a gamma spectrum and the determination of the ^{137}Cs, ^{134}Cs, ^{154}Eu content, where ^{137}Cs and the ratios ^{134}Cs/^{137}Cs and ^{154}Eu/^{137}Cs have specific correlations to the burnup of the irradiated fuel assembly under consideration.

Another method consists in the measurement of the neutron emission coming from an irradiated fuel element as it is done when using the Fork detector [1,2].

2.1. Physics principles

The neutron emission of a spent fuel element is mainly due to presence of ^{242}Cm and ^{244}Cm, which are essentially produced by successive neutron capture in ^{238}U, and the trans-uranium chain that follows from it [3].
The neutron emission due to ^{244}Cm at the discharge date is related to the burnup *Bu* by a power function [2], above 10 GWd/tU:

$$n_{244Cm}(Bu,0) = a \times Bu^{b} \tag{1}$$

The response of the Fork detector includes all the neutrons emitted by the fuel at the time *t* after discharge. Moreover the neutrons emitted by the SF undergo a certain multiplication due to the residual presence of fissile material and attenuation due to the presence of material in the detection geometry. These effects are a function of the measurement geometry and composition and so also of the burnup. We can therefore write the measured count rate *C* as:

$$C(Bu,t) = m(Bu,t) \times n(Bu,t) \tag{2}$$

The coefficient *m* includes detection efficiency and neutron multiplication factor which in principle depends on the composition of the fuel and the detection geometry and composition.

Indicating with F_1 the correction for the ^{244}Cm decay and with F_2 the share of ^{244}Cm on the total emitted neutrons at the measurement time, one can express the count rate due to ^{244}Cm at the discharge date as,

$$C_{244Cm}(Bu,0) = C(Bu,t) \times F_1(t) \times F_2(Bu,t) \tag{3}$$

Calculations based on the Origen-ARP code indicate that F_2 is a function of burnup and cooling time and between 0.95 and 0.98 for a cooling time *t* between 3 and 15 years and a burnup between 35 and 55 GWd/tU.
The neutron emission due to ^{244}Cm at the discharge date is then,

$$n_{244Cm}(Bu,0) = a \times Bu^{b} = \frac{C_{244Cm}(Bu,0)}{m(Bu,0)} \tag{4}$$

and assuming that the component due to the multiplication can be included in the power function one simply writes,

$$C_{244Cm}(Bu,0) = a' \times Bu^{b'} \tag{5}$$

The coefficients *a* and *b* can be determined via dedicated burnup calculations, while the coefficients *a'* and *b'* contain a combination of experimental data as detection efficiency and could be determined by coupling the fuel composition obtained from burnup calculations with a transport code like MCNPX.

The coefficients *a'* and *b*' depend on:
- the fuel type (MOX, LEU, initial enrichment)
- the measurement conditions (amount of boron dissolved in water, geometry, materials)

In this paper, the analysis of the neutron data consists in the determination of the coefficients *a'* and *b'* from measurements of fuel elements with known burnup. The data are grouped according to the initial enrichment of the fuel element. The influence of the initial enrichment is due to the fact that the burnup of an assembly is proportional to the integral of neutron flux and the total fissile material mass in the assembly. This means that from two assemblies that were submitted to the same fluence, the one with the highest initial enrichment obtained the highest burnup, or otherwise said, from two assemblies with the same burnup, the one with the highest initial enrichment will have the lowest neutron emission rate. This is also confirmed by experience [7].

2.2. Fork detector in the NPP of Doel

The Fork detector measures the neutron emission and the gross gamma emission from a submerged fuel assembly. The Fork detector is a watertight High Density Polyethylene (HDPE) detector head containing two sets of ionization chambers and fission chambers for measuring opposite sides of the fuel assembly simultaneously. Each arm contains an ionization chamber operating in current mode to measure the total gamma-ray output and one fission chamber operating in pulse mode to measure the total neutron output. The HDPE body is wrapped by a cadmium layer so that only the epithermal neutrons are measured.

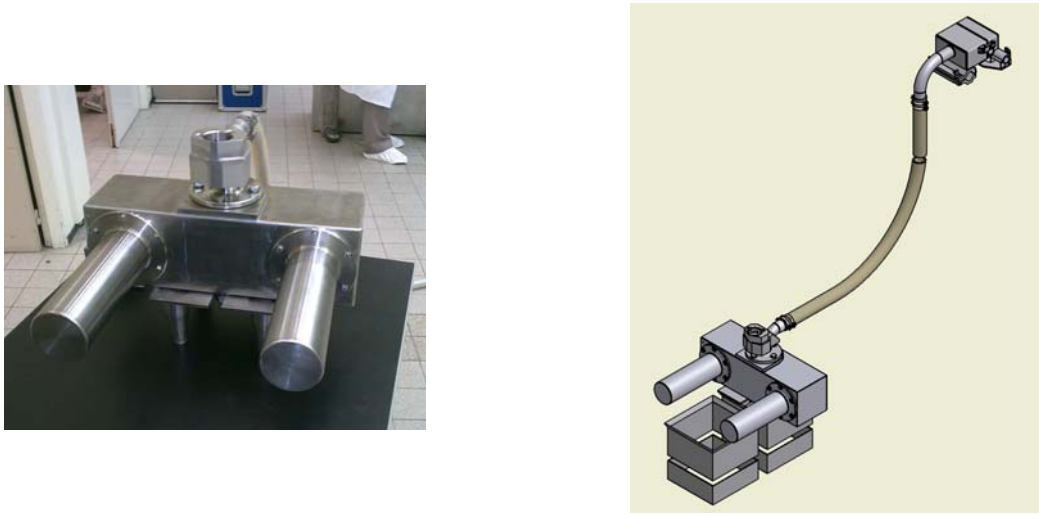

Figure 1: A picture and a technical drawing of the Fork detector.

The Fork detector (Figure 1) consists of a submerged structure, composed of a stainless steel housing, a flexible connection tube, electronic equipment, and a system for data acquisition.

The stainless steel housing is mounted on a stainless steel support, made compatible with the cell dimensions of the storage racks in use at the NPP of Doel; the support is installed in one cell while the housing is adjusted over the adjacent cell, in which the fuel can be lowered and prepared for the axial scan of the FE. The housing is adjusted over the fuel for optimal neutron and gamma signal.

The link between detector and electronics is assured by cables that pass through a silicone flexible tube that keeps the system watertight. The cables end in an interim box installed at the margin of the spent fuel pond, from which the connection can be made to the electronics rack, for signal handling and processing.

The combined housing and support are installed on the rack by using a long tool (made in 2 pieces to allow proper handling of the system) at every measurement campaign. The support is supposed to remain fixed to the Fork body, while a long tool is made installation specific.

The fuel is handled by a Fuel Handling Machine (FHM), equipped with a system allowing a precise positioning in the spent fuel pond; this system is used to carry out the axial scan of the fuel assembly. For each measurement, the axial position is read at the FHM and given as input to the data acquisition system.

The concept is adapted to assure a quick and safe transport of the installation, and easy and rapid connection after moving the equipment. The materials used allow easy decontamination to transport all equipment to another unit of the site.

The operation of the data acquisition system is managed by a computer (PC), which is connected to the electronic components and provides data processing, but does not affect the movement of spent fuel. The data acquisition system is installed on the PC and manages the interactions with the electronic modules, carries out the data acquisition, and saves the measurement results. The data treatment as outlined in the previous paragraph is automatically carried out and the burnup is determined in the mid point position and at the extremities of the FE. The measurement points and calibration curves are predefined by the user and can be customized. The saved data can be displayed and the burnup can be recalculated in case one wants to use a different calibration curve. The system generates also certificates and reports to be exported in document and PDF format.

3. Calibration measurements results

To determine the calibration parameters *a'* and *b'*, the neutron emissions are measured for some assemblies of the same initial enrichment, and a variety in burnup and cooling time.
A total of 22 SFEs were measured during the calibration measurement campaign. The characteristics (declared mean burnup, initial enrichment and cooling time) of the measured fuel element are summarized in Table 1. The active fuel length was either about 3.6 m (D3) or 4.2 m (D4). Some of the fuel elements contained Gd as burnable neutron poison. The amount of Gd is limited and is expected to be burnt after one year of irradiation; its presence in the FE should not affect the determination of the calibration curve. The nominal initial enrichments are limited to 4.25%, 4.35% and 4.45%. Fuel with lower uranium enrichments has no obligation for burnup verification by using a Fork measurement.

The SFE was positioned with a FHM equipped with device to determine the SFE height. Three fine 10 cm step axial scans were carried out in order to verify the shape of the burnup profiles. The position of the SFE was determined by the readings of the FHM and the beginning and the end of the active fuel length could be determined.
A measurement time of 60 s per point was sufficient to reduce the uncertainty due to statistics below 1%.

The calibration was carried out using the mid-point measured count rate corrected for ^{244}Cm decay as in equation (3). This quantity was compared with the mid-point burnup which is obtained from the operator declared value and the ratio between the mid point burnup and mean burnup. The mid-point burnup is higher than the mean declared burnup and therefore the ratio is larger than unity. The ratio can be obtained from the measured profiles. It is clear that the ratio was determined with an iterative procedure as the calibration curve coefficients are not known a priori and they are needed to obtain the burnup profile from the measured neutron count rate profile.

Using the neutron data from the full scanning of the fuel element D3V16 the ratio between mid-point burnup and mean burnup is 1.094. This value is kept constant through the fuel elements 3.6 m long.
Using the neutron data from the full scanning of the fuel element D4Q14 the ratio between mid-point burnup and mean burnup is 1.068. This value is kept constant through the fuel elements 4.2 m long.
An example of neutron count rate profile is shown in Figure 2.

Fuel Name	Initial Enrichment (%)	Cooling Time (d)	Declared Mid-point Burnup (MWd/t)	Measured Mid-point Burnup (MWd/t)	Difference %
D3V16	4.2516	2813	57164	57012	-0.27%
D3V41	4.2545	3141	53092	53240	0.28%
D3V35	4.2486	3488	47665	47327	-0.71%
D3V03	4.276	3488	50667	50677	0.02%
D3Z02	4.4547	1616	58894	58850	-0.07%
D3C16	4.4508	880	53055	53762	1.33%
D3Y03	4.4464	2421	47477	47380	-0.20%
D3Y22	4.443	2421	50635	50659	0.05%
D3X32	4.436	2813	50240	50355	0.23%
D4Y10	4.2487	502	46225	48226	4.33%
D4Q14	4.2592	2361	49387	49407	0.04%
D4Y05	4.2498	500	55640	56465	1.48%
D4Q01	4.2500	3143	47131	47549	0.89%
D4Q03	4.2500	2362	49261	49253	-0.02%
D4Q36(*)	4.2500	2361	40621	40455	-0.41%
D4Q35(*)	4.2500	4597	15588	11234	-27.93%
D4Q37(*)	4.2500	1559	36836	36892	0.15%
D4V04	4.3529	1558	43210	43263	0.12%
D4X04	4.3461	1039	49036	50421	2.82%
D4U13	4.3509	2013	53572	53615	0.08%
D4U16	4.3500	2362	44586	44022	-1.26%
D4V19(*)	4.3500	2013	44113	44566	1.03%

Table 1: Spent Fuel elements. The fuel elements whose name ends with (*) contained Gd as burnable poison.

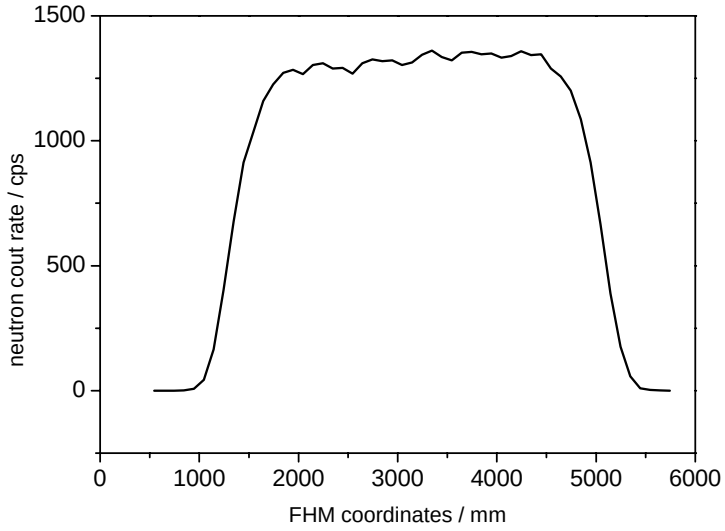

Figure 2: Example of a neutron count rate profile.

The fitting of the data was done using the Deming code [8]. Only measurement data with a minimum cooling time of 3 years and minimum declared burnup of 30 GWd/tU were considered. In this way the correction factor F_2 can be considered constant and included in the parameter *a'* of Equation (5).
The following independent uncertainties were considered in the fitting procedure:

- 1% on the declared mean burnup
- 1% on the ratio mid point to mean burnup
- Statistical uncertainty on the neutron count rate
- 0.5% on the F_2 factor

The parameters of the calibration curves for the various fuels are given in Table 2. The uncertainty on the parameters and their correlation coefficients are also given. The data corresponding to SFEs with a 4.25% initial enrichment coming from both D3 and D4 reactors could be fitted with one calibration curve.

Initial Enrichment	a'	b'	$\sigma_{a'}$	$\sigma_{b'}$	$\sigma^2_{a'b'}/\sigma_{a'}\sigma_{b'}$
4.25%	11.43×10^{-4}	3.645	2.06×10^{-4}	0.047	-0.99947
4.35%	5.76×10^{-4}	3.796	5.59×10^{-4}	0.253	-0.99975
4.45%	15.29×10^{-4}	3.551	3.03×10^{-4}	0.050	-0.99979

Table 2: Resulting coefficients from the data analysis.

The high uncertainties on the parameters for the 4.35% SFEs are a consequence of a non perfect fit of the data. To calculate the statistical uncertainty due to the uncertainty on the fit parameters on the measured burnup, one has to account for the correlation between the parameters. It was verified that the maximum uncertainty on the measured burnup, accounting for the full covariance on the parameters, was less than 0.6 GWd/tU for the 4.35% SFEs data set. The maximum difference between declared and measured burnup was 1.3% for the data with cooling time of at least 3 years.

In general, the differences between declared and assessed values give an idea about the accuracy of the approach, and its validity. However, it should be noted that the approach used results in a consistency check as it relies entirely on the declared burnup used in the calibration measurement.

The measured burnup was however determined for all the measured SFEs and reported in Table 1. The F_2 factor for SFEs used in the calibration procedure is included in the parameter *a'* of Equation (5). For the SFEs with burnup lower than 30 GWd/tU or cooling time less than 3 years, the measured burnup was determined accounting for the correction factor F_2 relative to the F_2 factor for SFEs used in the calibration procedure. The correction factor F_2 was determined with calculations based on the Origen-ARP code [9] using the declared irradiation history for the SFEs.

The good agreement between measured and declared burnup is an indication that that the F_2 factor is approximately constant on condition that the cooling time is between 3 and 15 years and the burnup between 35 and 55 GWd/tU. For shorter cooling times, despite accounting for the F_2 factor, the measured mid-point burnup is systematically higher than the declared mid-point burnup and the observed difference is always less than 5%. The obtained calibration curve significantly underestimates the burnup for the FE D4Q35 by 28%; this large discrepancy may indicate that the model in use is not sufficiently reliable for SFEs with low burnup and loaded with Gd. More investigations are needed to explain this discrepancy.

The burnup in the first and last 50 cm of the fuel was also assessed, by measuring at 0, 25 and 50 cm distance from the extremities of the fuel active length. The burnup is not completely linear in the last 50 cm. For example for the FE D3V16 the mean value is 41.8 and 42.3 GWd/tU for the first and last 50 cm, respectively. The corresponding point values at 25 cm from the ends are 46.1 and 46.7 GWd/tU respectively. The average of the values at 0, 25 and 50 cm from the ends are taken as representative for the mean burnup of the extremities.

4. Conclusions

A refurbished Fork detector measurement system has been installed at the NPP of Doel (Belgium). A new mechanical tool and rack support for the installation and fixation of the Fork detector in the pond and a user-friendly software for the data acquisition and analysis were developed. The equipment is capable of measuring all assemblies, independent of burnup, cooling time and initial enrichment.

Calibration curves were obtained for the requested enrichments 4.25%, 4.35% and 4.45%, based on the measurement of available fuel assemblies.

Based on the results of the measurements carried out, the SFEs loaded with Gd yielded a quasi identical neutron count rate when compared with SFEs without Gd and with comparable burnup. This is expected since the Gd is burned after one year of normal operation of the reactor. Therefore there was no need for a separate calibration curve for the SFEs loaded with Gd.

Based on the calibration measurements, the measured burnup corresponds to the declared burnup within 2% if the cooling time is at least 3 years and the burnup is between 30 and 55 GWd/tU. The deviation increases outside these validity ranges. In particular, it was verified that the obtained calibration curve underestimates by 27% the burnup for the FE D4Q35 for which a 15.6 GWd/tU mid-point burnup was declared.

The measured values at the extremities of the SFEs are used to assess the burnup in the first and last 50 cm.

Adding more measurement points to the calibration curve will improve the statistics and possibly extend its validity range.

5. Acknowledgements

The authors would like to thank the operators of the fuel team for their technical support during the measurements at the NPP of Doel (Belgium).

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06 Destructive Analysis – Quality

Preparation and development of new Pu spike isotopic reference materials at IRMM

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Abstract:

Reliable isotope measurements of nuclear material and reference materials with small uncertainties of the certified values are of great importance for security and safeguards of nuclear installations. They provide the basis for a strong verification and detection system to safeguard nuclear activities. Worldwide needs for continued and improved Isotopic Reference Materials (IRM) are the main reason for developments of new nuclear reference materials at IRMM. Measurement capabilities of laboratories have evolved considerably over the years hand in hand with progress in modern analytical techniques. But some plutonium reference materials are already on the market for decades. The availability of new reference materials with appropriate small uncertainties of the certified values is essential for measurement laboratories striving to meet the International Target Values for Measurement Uncertainties in Safeguarding Nuclear Materials (ITVs). The preparation and the certification of such materials are demanding and challenging tasks that require state-of-the-art measurement procedures and equipment. The Institute for Reference Materials and Measurements (IRMM) has repeatedly demonstrated its capabilities in plutonium analysis and represents one of the few institutes that supplies plutonium IRMs worldwide. IRMM has recently performed an inter-calibration campaign inter-linking selected plutonium spike IRMs on a metrological basis, applying state-of-the art measurement procedures. In the scope of this compatibility study new reference materials have been prepared for Isotope Dilution Mass Spectrometry (IDMS) in nuclear fuel cycle measurements.

A new series of large-sized dried (LSD) spikes IRMM-1027n has been prepared and certified for plutonium and uranium amount content and isotopic composition. These mixed spikes are applied to measure the uranium and plutonium content of dissolved fuel solutions using isotope dilution mass spectrometry. They are prepared by IRMM to fulfil the existing requirements for reliable spike IRMs in fissile material control from European Safeguards authorities and customers from industry.

IRMM-046b, a mixed uranium-plutonium spike IRM of highly enriched ^{233}U and ^{242}Pu that dates from 1995 was recertified for isotope amount content and isotopic composition, each with considerably smaller combined uncertainties. IRMM-046c, a new mixed uranium-plutonium spike and IRMM-049d, a highly enriched ^{242}Pu spike have been prepared and certified. IRMM-049d was prepared from the same stock solution as its predecessor IRMM-049c, dating from 1996, but this ^{242}Pu spike has certified values with smaller combined uncertainties. These reference materials were prepared by dissolving the base materials in nitric acid and are certified for the amount content by gravimetry or isotope dilution mass spectrometry (IDMS). The traceability of the certified values to the SI is established through an unbroken chain of comparisons all having stated uncertainties.

IRMM is also cooperating with the Institute for Transuranium Elements (EC-JRC-ITU) in a feasibility study on the development of Pu reference materials for "age dating" in nuclear forensics. In the course of this work the reference materials NBS SRM 946, 947 and 948 (NBL CRM 136, 137 and 138) will be investigated among others.

Keywords: plutonium; spike isotopic reference materials; IDMS, traceability.

1. Introduction

Confidence in comparability and reliability of measurement results in nuclear material analysis is established via reference materials, reference measurements and inter-laboratory comparisons. They provide the basis for a strong verification and detection system to safeguard nuclear activities in line with the Treaty on the non-proliferation of nuclear weapons (NPT) and the Additional Protocol (AD).
The Institute for Reference Materials and Measurements (IRMM) is one of the leading institutes worldwide that develops and certifies nuclear reference materials to fulfil the existing requirements for reliable certified reference materials (CRMs) in fissile material control. Worldwide needs and advancements in analytical techniques over the last decade have led to more stringent requirements for laboratory performance in nuclear material accountancy. The International Target Values for Measurement Uncertainties in Safeguarding Nuclear Materials (ITVs) are uncertainties to be considered in judging the reliability of analytical techniques applied to industrial nuclear and fissile material that are subject to safeguards' verification. The IAEA took over the concept of ITVs in the early 1990's from the ESARDA Working Group of Standards and Techniques for Destructive Analysis (WGDA). During 2010 the ITVs were revisited by the IAEA, ESARDA, INMM and other expert groups and published as ITV2010 in November 2010 [IAEA-SRT-368]. They are intended to be used by plant operators and safeguards organizations, as a reference of the quality of state-of-practice measurements achievable in nuclear material accountancy. Therefore, development of particular plutonium and uranium reference materials with the smallest possible uncertainties of the certified values are needed in nuclear security and safeguards in line with advancements of analytical techniques.
A new certificate for IRMM-046b, a mixed uranium-plutonium spike of highly enriched ^{233}U and ^{242}Pu was issued in 2010 for isotope amount content and isotopic composition, each with considerably smaller combined uncertainties than in the previous certificate. IRMM CRMs used for isotope dilution mass spectrometry were approaching exhaustion. To guarantee future provision of these valuable materials to the nuclear measurement community it was decided to replace them by preparing two new spike reference materials, IRMM-046c and IRMM-049d. The certification of these new CRMs was part of an IRMM compatibility study interlinking various plutonium spike CRMs on a metrological basis [1].
A new batch of the IRMM-1027 large-sized dried (LSD) spike series was prepared. This isotopic reference material series is designed for fissile material control at the Euratom Safeguards on site-laboratories. These spikes are primarily used for the determination of Pu and U amount content by IDMS in spent fuel solutions at the reprocessing plants at La Hague and Selafield. The preparation and the certification of the new batch IRMM-1027n are discussed in more detail in the certification report [2].
IRMM is also engaged in a feasibility study for the development of plutonium reference materials for age dating to be used for method validation purposes in nuclear forensics applications. The "age" of a nuclear material is defined as the time that has passed since the last chemical separation of the mother and daughter isotopes. Different "clocks" (pairs of mother and daughter radionuclides) can be used for the determination of the unknown age of a material. In case of plutonium the isotope pairs $^{241}Pu/^{241}Am$, $^{238}Pu/^{234}U$, $^{239}Pu/^{235}U$, $^{240}Pu/^{236}U$, and possibly $^{242}Pu/^{238}U$ can be used as "clocks". Some preliminary results for NBS SRM 946 will be presented here.

2. Experimental

2.1. Preparation of IRMM-1027n

High purity metals were chosen as starting materials for the IRMM-1027 LSD series. Plutonium MP2 metal (^{239}Pu) from Cetama, natural uranium (EC NRM 101) and highly enriched ^{235}U metal (CRM-116) were dissolved in concentrated nitric acid in a 3 L long-necked borosilicate flask. All weighings were carried out accurately, with reference to a set of calibrated weights traceable to the SI.
Approximately 1200 units were dispensed into penicillin vials by a validated automated system. The solution in each vial was dried down and then covered with a light layer of an organic polymer, cellulose acetate butyrate (CAB), as stabiliser during storage and transport [2, 3].

2.2. Preparation of IRMM-049d and IRMM-046c

For the preparation of the IRMM-049d spike reference material, a stock solution was made by dissolving ^{242}Pu metal in 5 mol $\cdot L^{-1}$ nitric acid. This stock solution was purified from the daughter

products and other impurities by anion exchange. The eluted Pu fraction was evaporated to dryness and dissolved in 5 mol $\cdot L^{-1}$ nitric acid to obtain a 10 mg Pu per g solution. From that purified Pu solution, a fraction was taken and diluted with 5 mol $\cdot L^{-1}$ nitric acid to obtain the final concentration of ^{242}Pu of 0.1 mg Pu per g solution.

For the preparation of the mixed plutonium-uranium IRMM-046c spike, ^{233}U metal was dissolved in 8 mol $\cdot L^{-1}$ nitric acid and purified by anion exchange. The uranium fraction was eluted with 8 mol $\cdot L^{-1}$ nitric acid and evaporated to dryness. A fraction of a 10 mg Pu per g solution, already used for the preparation of IRMM-049d, was added to uranium and diluted with 8 mol $\cdot L^{-1}$ nitric acid. The final concentration of ^{242}Pu was 0.1 mg Pu per g solution and of ^{233}U 1 mg per g solution.

Different steps of the preparation of IRMM-049d and IRMM-046c isotopic reference materials are shown in Figure 1.

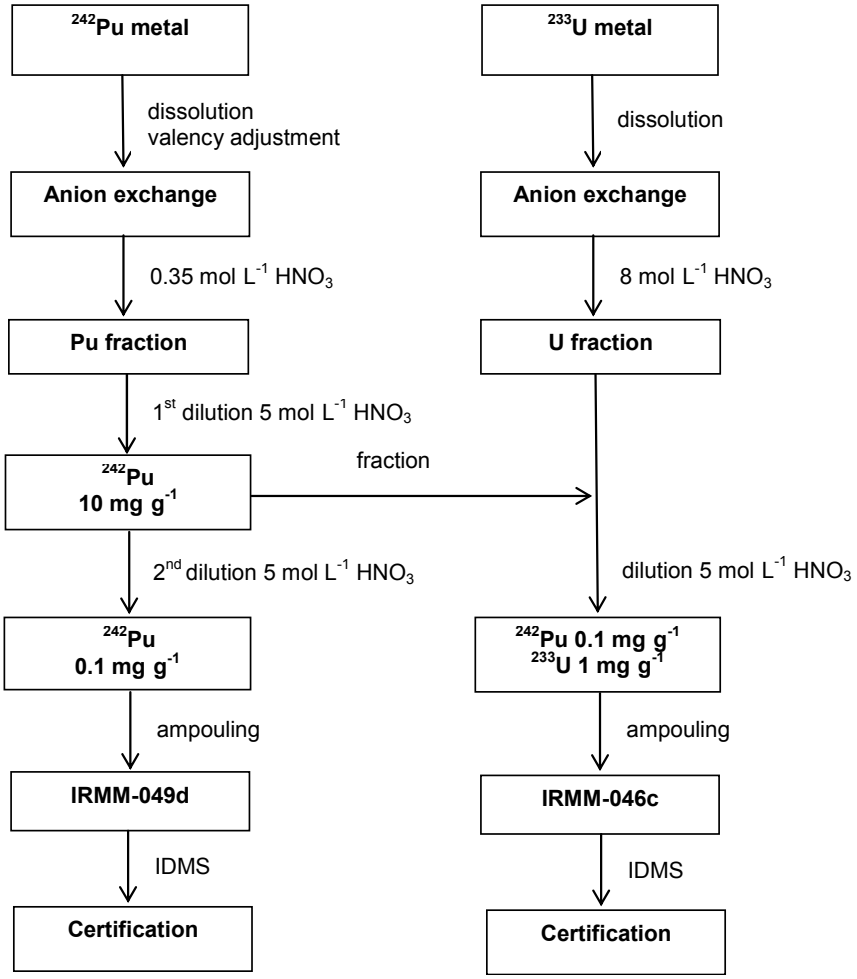

Fig.1 The flowchart for the preparation of IRMM-049d and IRMM-046c.

2.3. Isotope measurements by Thermal Ionisation Mass Spectrometry (TIMS)

Isotope Dilution Mass Spectrometry (IDMS) was applied for the measurements of the plutonium and uranium amount contents. This is a reliable analytical technique and widely used in nuclear safeguards, especially when high quality results with small measurement uncertainties are needed.
Prior to mass spectrometry a chemical procedure using anion exchange was applied [1]. The purified fractions of U and Pu were prepared in 1 mol $\cdot L^{-1}$ nitric acid and loaded on a Re filament. The isotopic ratios of uranium and plutonium were measured on a Triton TIMS using the total evaporation technique. Applying the total evaporation technique the measurement is continued until the sample is exhausted. This is done in order to minimize mass fractionation effects. The mass spectrometers were

calibrated for mass fractionation by measuring IRMM-074/10 uranium and IRMM-290A/3 plutonium isotopic reference materials [4, 5].

3. Results and Discussion

3.1 IRMM-1027n

A new set of IRMM-1027n LSD spikes, containing about 50 mg of uranium with a ^{235}U mole fraction of 19.5 % and about 1.8 mg of plutonium with a ^{239}Pu mole fraction of 97.8 % was prepared and certified for Pu and U isotope content and isotopic abundance. The amount content of the spikes is such that no dilution of a typical sample of dissolved fuel is needed before the measurement by IDMS using a single LSD spike. The certification of the 1027 LSD series is done via gravimetry. In addition the certified values are verified by IDMS. With this approach IRMM provides high-quality isotopic reference materials to the nuclear measurement community applying two independent “primary” methods for certification and verification, underpinning the confidence in the certified values. The U amount content of 1027n was therefore certified based on the values from mass metrology of the validated automated system. From the eleven measurements used to asses the homogeneity of the whole series (1200 units), four were selected at random to verify the U amount content by IDMS [2].
The results of the verification measurements are shown in Figure 2. The IDMS measurement results agreed well with the values for uranium amount content calculated from the amounts of dissolved metals and solution.

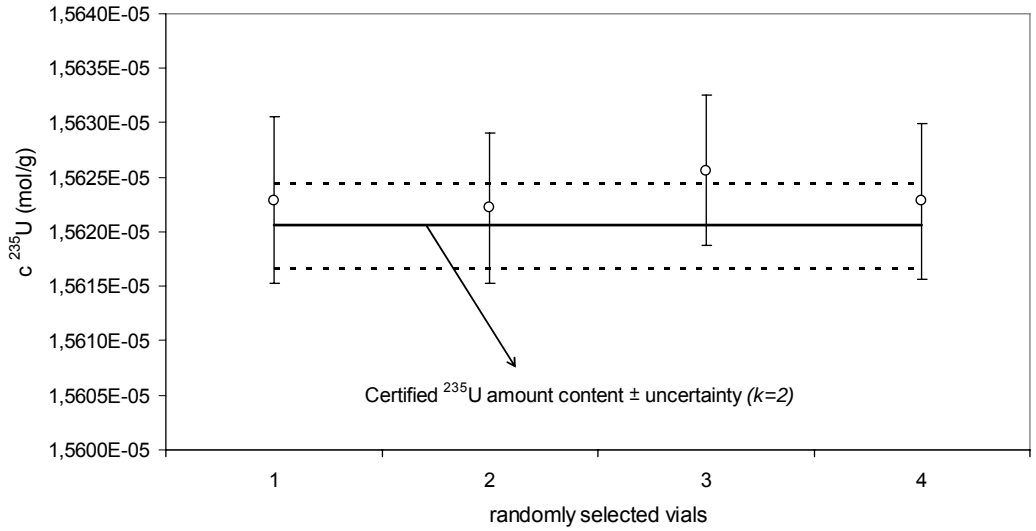

Fig.2 Amount content of ^{235}U in IRMM-1027n (from the masses of metals and solution) compared with the measured values by IDMS (with expanded uncertainties, *k=2*).

The situation was different in the case of plutonium. After applying the same procedure during preparation of the stock solution as in previous batches of the 1027 solution it was found that the Pu metal did not dissolve completely. A fine white deposit was observed even after several weeks of a continuous dissolution process. Taking into account the limited supply of plutonium metal and that IRMM has a long record of demonstrated measurement capabilities in plutonium analysis; it was decided not to discard the batch solution. The uranium metals were added to the solution and after homogenisation filtered through a separation column [1]. Due to this incomplete dissolution of the MP2 Pu metal a deviation of about -1.2 % from the gravimetric value for the Pu amount content was observed. It was decided to certify the plutonium amount content by IDMS applying Thermal Ionisation Mass Spectrometry. IDMS can be regarded in this case as "primary" measurement method which has proven to provide accurate results also for previous batches of the IRMM 1027 series. The recently certified uranium-plutonium IRMM-046b CRM was used as a spike for the IDMS linking the certification for Pu amount content of 1027n to the IRMM compatibility study on selected Pu spikes [1]. The certified value for the ^{239}Pu amount content was calculated as the mean value of the certification

measurements and is 2.791 3 (12) 10^{-6} mol $\cdot g^{-1}$ [2]. The results for the plutonium amount content are also traceable to the SI via MP2 but with two additional steps in the traceability chain. IRMM-1027n - via IRMM-046b - via IRMM-1027m (and verified by Eqrain-11) - via MP2 to SI [1]. In the frame of the ongoing support task EC A 1806 *Verification of mixed U-Pu Spikes* between IRMM, IAEA and ITU additional verification measurements of 1027n will be available in the near future.
The individual IDMS results together with the certified value are shown in Figure 3.

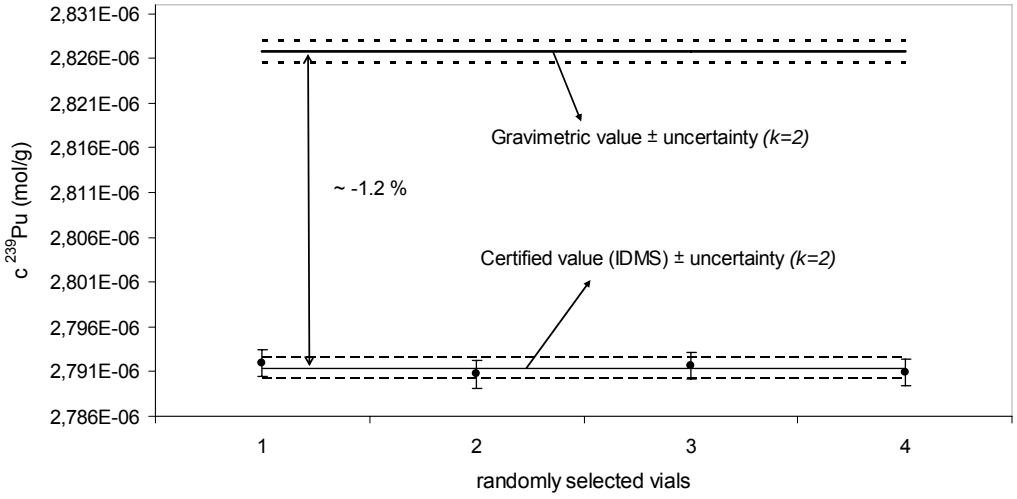

Fig.3 Amount content of ^{239}Pu in IRMM-1027n (from the masses of metals and solution) compared with the measured values by IDMS (with expanded uncertainties, *k=2*).

3.2 IRMM-049d, IRMM-046b and IRMM-046c

IRMM produces and maintains solutions of enriched uranium and plutonium isotopes designed for mass-spectrometric isotope dilution measurements of nuclear materials. They are part of a systematic IRMM programme to supply various nuclear reference materials at different concentrations. For the measurement of plutonium, the isotope ^{242}Pu is valuable as a spike because this isotope is usually found only as a minor component of plutonium in the nuclear fuel cycle.

A new certificate for IRMM-046b, a mixed uranium-plutonium spike of highly enriched ^{233}U and ^{242}Pu was issued in 2010 for isotope amount content and isotopic composition, each with considerably smaller combined uncertainties than in the previous certificate.
To replace exhausted stocks of CRMs used for isotope dilution mass spectrometry, IRMM prepared and certified IRMM-046c, a new mixed uranium-plutonium spike and IRMM-049d, a highly enriched ^{242}Pu spike. Each of these new Pu CRMs has a similar certified value as its predecessor but with considerably smaller combined uncertainties. Advancements and development of state-of-the-art mass spectrometric techniques and instrumentation over the years resulted in reduction in measurement uncertainty by a factor of 3 or more. Table 1 illustrates this achievement comparing the relative expanded uncertainties of various IRMM CRMs.

		Rel Uc *(k=2)* in %	
	Year of certification	^{242}Pu	^{233}U
old IRMM-046b	1995	0.15	0.15
recertified IRMM-046b	2010	0.039	0.021
IRMM-046c (indicative values)	2011 ongoing		
IRMM-049 (exhausted)	1989	0.15	
IRMM-049c	1996	0.13	
IRMM-049b	1998	0.067	
IRMM-049d	2011	0.046	

Table 1 Relative expanded uncertainties of selected spike isotopic reference materials.

3.3 External plutonium interlaboratory comparison programme Eqrain-11

Eqrain is the inter-laboratory comparisons programme organised at regular intervals for the analysis of uranium and plutonium by CETAMA (Commission d'ETAblissement des Méthodes d'Analyse du CEA) [6]. The certified test samples of Eqrain-11 consisted of three plutonium nitrate solutions (M29, M57 and M107) with undisclosed values for Pu amount content.
IRMM linked the participation in Eqrain-11 to an inter-calibration campaign by determining the plutonium isotope content applying IDMS using various selected spikes and to the certification of IRMM-046b, IRMM-046c and IRMM-049d. The aim of the campaign was to check the quality of various selected spikes and to demonstrate IRMM`s measurement capabilities for plutonium measurement via external quality tools. The results reported by IRMM were in excellent agreement with the reference value provided by CETAMA [1]. Eqrain-11 is still ongoing; therefore the plutonium amount content and the reference values are not disclosed in this paper. As an example, the IDMS measurement results for M107 sample normalised to the Eqrain-11 reference value are shown in Figure 4.

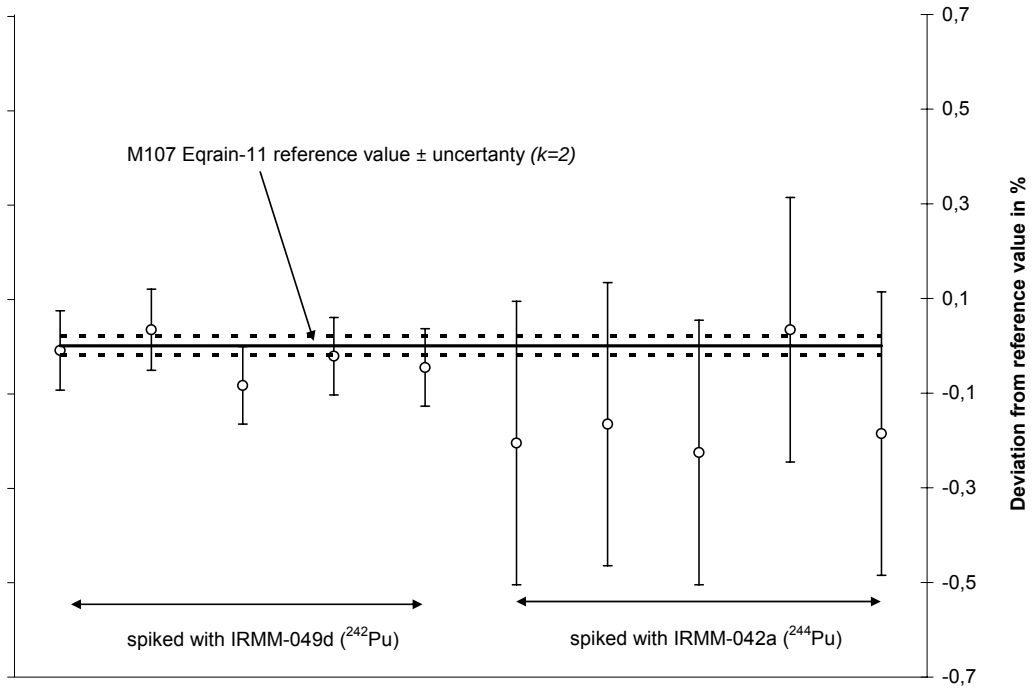

Fig.4 Normalised amount content of ^{239}Pu in Eqrain-11 M107 sample compared with the measured values by IDMS (with expanded uncertainties, *k=2*).

3.4 Preliminary results of NBS SRN-946 for "age" dating

To resolve the current lack of nuclear reference materials certified for their separation date needed in nuclear forensics, several plutonium materials with different isotopic compositions and production dates are being characterized at IRMM in course of a feasibility study for reference materials for nuclear age dating. The results for three of the plutonium-uranium "clocks" for NBS SRM 946 – a plutonium reference material certified for isotopic composition – are shown in Figure 5. These measurements were performed by TIMS applying IDMS. The ages derived from these three different mother-daughter isotope systems ($^{238}Pu/^{234}U$, $^{239}Pu/^{235}U$ and $^{240}Pu/^{236}U$) do not differ significantly from each other.

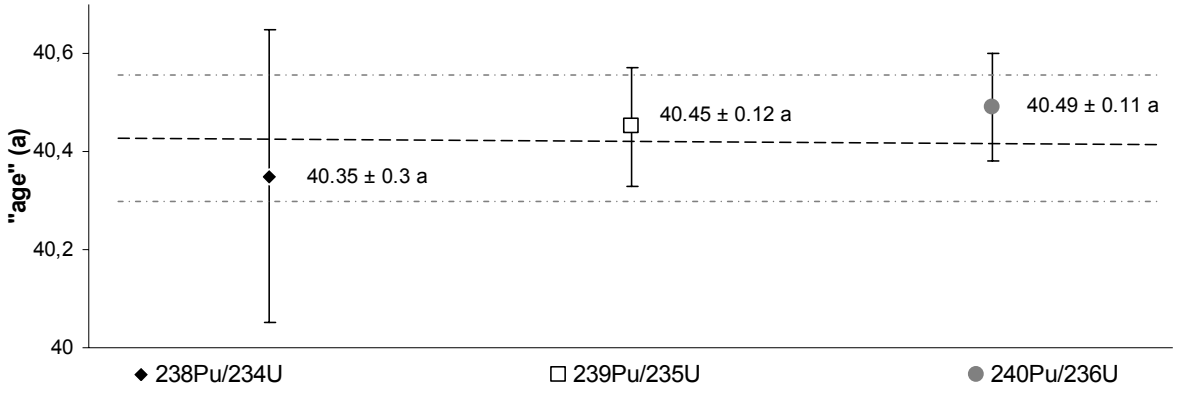

Fig.5 Age in years of NBS SRM 946 (calculated for 17 October 2010) for the "clocks" $^{238}Pu/^{234}U$, $^{239}Pu/^{235}U$ and $^{240}Pu/^{236}U$ with expanded uncertainties *(k=2)*.

4. Conclusions

The prime objective of the IRMM is to build confidence in the comparability of measurements by the production and dissemination of internationally accepted quality assurance tools, including high-quality certified reference materials. A new series of LSD spikes for IDMS determinations of uranium and plutonium content in solutions of spent nuclear fuel from reprocessing plants has been prepared. The uranium content was certified based on gravimetry and successfully verified by IDMS on individual vials. Plutonium was certified by IDMS via a recently recertified IRMM-046b spike reference material. New isotopic reference materials, IRMM-046b, IRMM-046c, mixed uranium-plutonium and IRMM-049d, highly enriched in ^{242}Pu were prepared and certified for amount content by IDMS. These materials were prepared in the frame of IRMM`s programme to supply various spike isotopic reference materials at different concentrations to the nuclear safeguards and nuclear material measurement community. Furthermore, IRMM has successfully demonstrated Pu measurement capabilities via external quality tools by participation in Eqrain-11 applying IDMS and using various selected Pu spikes reference materials. With the development of new plutonium spike isotopic reference materials IRMM significantly contributes to the availability of these materials in the future. IRMM is regularly exchanging views with the customers and users of Pu reference materials on further needs and developments.

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IAEA On-Site Laboratory: Experience in establishing an internal verification regime based on method inter-comparison with ID/TIMS

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Abstract:

Within the scope of IAEA's safeguards approach for the Rokkasho Reprocessing Plant (RRP) in Japan, a number of analytical techniques are used in the On Site Laboratory (OSL) to meet the high demands on analytical chemistry related to nuclear material accountancy. Among those techniques, thermal ionization mass spectrometry combined with isotope dilution (ID/TIMS) is used as a Quality Control reference for all other U and Pu assay methods, and serves as an internal verification tool to support the credibility of OSL results. The older of the two TIMS instruments has operated reliably for more than six years. In view of the increasing number of samples foreseen, the age of the first instrument, and to better support timely reporting of analytical results, a second TIMS was installed in 2010. Successful acceptance testing and subsequent internal and external inter-comparison exercises have demonstrated the suitability of the second TIMS at OSL as a reliable internal quality reference measurement system.

Key words: Nuclear Safeguards, TIMS, Isotope Dilution, Quality Control

1. Introduction

The establishment of the jointly operated On-Site-Laboratory (OSL) at the Rokkasho Reprocessing Plant (RRP) in Japan entails a number of advantages for both the IAEA Inspectorate and Japanese authorities. Among the most important objectives are the timely provision of analytical results and the avoidance of costly overseas nuclear sample shipment. A team of designated IAEA Safeguards Inspectors and the Japanese NMCC Analysts (Nuclear Material Control Centre) are the two counterparts cooperating in OSL. Among other duties, NMCC has the overall responsibility for the operation and management of the OSL in contract with JSGO (Japanese Safeguards Organization) under the auspices of the Ministry of Education, Culture, Sports, Science and Technology (MEXT).

Approximately 400 samples per year are to be analyzed in OSL when full scale reprocessing operations are started at RRP. The sample diversity in terms of plutonium concentration and U/Pu ratio with high α or $\beta\gamma$ activity levels necessitates the employment of adequate destructive and non-destructive analytical methods to meet safeguards requirements. The choice of the analytical method is mainly driven by the sample composition and the analytical request by the IAEA's Inspectorate. At

the OSL, two hybrid k-edge densitometers (HKED) are installed, one connected to a hot cell line for highly radioactive spent fuel solutions and active waste samples, and the other connected to a glove box line for pure plutonium and uranium products and MOX samples. Spectrophotometry of Pu(VI), thermal ionization mass spectrometry (TIMS) and high resolution gamma spectrometry (HRGS) are additional analytical methods used in the OSL. Each method covers a different range of Pu concentration. A strategic role is ascribed to TIMS due to its high accuracy and high precision for element assay and isotopic abundances. Over the past seven years, continuous improvements were realized on the analytical procedures in OSL which led to a steady improvement of the performance of the first TIMS instrument (TRITON A) and to the development of the QC system. Essential progress has been achieved in the applied isotope dilution method (ID) and results have recently been presented elsewhere [1].

The key role of TIMS is also reflected in its function as a sound reference to control the performance of all other analytical instruments in the laboratory. Following established internal quality control principles, any result is subject to verification by a second analytical method [2]. This is achieved either by providing proof through ID/TIMS that equipment calibration is under control (e.g., the HKED systems) or by direct comparison of isotope ratios from TIMS and HRGS.

Both TIMS instruments at OSL are regularly used to measure samples from internal and external intercomparison exercises and internal inspections. Simultaneous measurements of the same standards on both instruments are carried out monthly and are deemed as a customary and appropriate measure within the scope of domestic verification methodology.

The most recent multi-collector TIMS at OSL (TRITON B) was installed beginning of 2010 and passed acceptance testing in September/October 2010. It now performs in compliance with International Target Values for measurement uncertainties [3]. Routine operation of TRITON B started in November 2010.

2. Experimental

The first thermal ionisation mass spectrometer (TRITON A) was installed in OSL in 2003 and tested in inactive mode with neodymium, went through a long examination process and started routine operations with uranium in 2005 and plutonium in 2006, after passing several acceptance test phases. In February 2010, the installation of the TRITON B was completed and underwent acceptance test in September/October of the same year. Both instruments are installed in a controlled environment with controlled temperature and humidity conditions and their ion source housings are attached to a low activity glove box line. The fully automated mass spectrometers are multi-collector instruments, respectively equipped with nine and seven Faraday cups, with a dynamic range of 0.1 mV to 50 V, and a secondary electron multiplier (SEM) without energy filter (RPQ). The total evaporation method is being utilized for routine sample analysis in combination with rhenium as ionisation filament and tungsten as evaporation filament. After U/Pu separation via column chromatography with Tri-Octyl-Phosphin-Oxide (TOPO), either 100 ng uranium or 50 ng plutonium are loaded onto a W filament for analysis, with up to 21 filaments per analysis sequence.

The measured ion current intensities are positioned at masses 233, 234, 235, 236 and 238 for uranium and 238, 239, 240, 241, 242 and 244 for plutonium. The certified reference materials (CRM) NBL U010 (U) and NBL 137 (Pu) are used to monitor the quality of routine measurements. To check the integrity of the above reference materials, periodical use of other certified solutions is made, such as NBL U500 (U) and NBL 144 (Pu).

On an annual basis, both TIMS instruments undergo an overall examination by the manufacturer's service engineer, as well as by a mass spectrometry specialist from the IAEA. If necessary, re-adjustment of experimental parameters according to QC and analytical requirements are carried out. Instrument stability over a wider range of isotope ratios is investigated at regular time intervals by using appropriate CRM such as NBL 030A, NBL U005A, NBL 128, and KRI 3568.

OSL has been participating since 2008 in international QC exercises such as the CETAMA EQRAIN programme for U analysis, domestic Pu round robin analyses with other Japanese laboratories and inter-comparison exercises with the IAEA Seibersdorf NML laboratory. The established performances

are in accordance with ITV 2010 (0.15%) for uranium assay. ANOVA evaluation of ID/TIMS results of OSL on concentrated Pu product solutions gave a random uncertainty component of 0.12% for Pu assay, which is below the ITV 2010 recommended value (0.15%).

3. Acceptance Test Results

In October 2010 the acceptance testing of TRITON B was completed. The results from reference materials used for the performance evaluation are summarized in Table 1. We focused on the n(U-235)/n(U-238) and n(Pu-240)/n(Pu-239) as they are of immediate interest to safeguards and have specified uncertainties in the International Target Values (ITV 2010) document. The average values are based on measurements of 15 filaments; observed K factors (i.e., measured divided by certified ratio) are close to 1. The relative deviations of the experimental average values versus respective certified value of the reference material are smaller than the certified uncertainty, hence the relative deviations from certified are not significant (i.e., bias is insignificant). Relative standard deviations on the main ratios are below 0.05% and considerably lower than respective ITV 2010 recommended values. For minor isotopes ratios (e.g., n(U-234)/n(U-238) satisfactorily low relative standard deviations were observed, although recommended values for these ratios are not specified in ITV 2010.

CRM	**U 500 (U)**	**U 010 (U)**	**KRI 3568 (U)**	**CRM137 (Pu)**	**CRM144 (Pu)**
Isotope ratio	235/238	235/238	235/238	240/239	240/239
Average (exp.)	0.999583	0.010137	1.000420	0.240869	15.10111
STDEV	0.00037	0.000005	0.000375	0.000008	0.0014
RSD	0.037%	0.046%	0.040%	0.003%	0.010%
ITV (RSD)	0.10%	0.20%	0.10%	0.10%	0.10%
Certified ratio	**0.999698**	**0.010140**	**1.000147**	**0.240884**	**15.10056**
Certified uncertainty	**0.1%**	**0.1%**	**0.061%**	**0.12%**	**0.045%**
Rel. Deviation	-0.011%	-0.029%	0.027%	-0.006%	0.004%
K factor	1.0001	1.0003	0.9997	1.0001	1.0000

Table 1. Results of Acceptance testing - TRITON B TIMS in OSL

Within the scope of the acceptance testing, loaded filaments with the same Pu and U standards were shipped to the IAEA Nuclear Material Laboratory (NML) in Seibersdorf, Austria for further measurements using a TRITON TIMS. NML results are tabulated in Table 2. The average values are the mean of 3 filaments with no outlier. The results of the OSL and NML are in good agreement. However, for both sets of measurements, one produced at OSL and one at NML, we observe a maximum relative difference of 0.03% to the certified value for all reference materials.

CRM	**U 500 (U)**	**U010 (U)**	**KRI3568 (U)**	**CRM137 (Pu)**	**CRM144 (Pu)**
Isotope ratio	235/238	235/238	235/238	240/239	240/239
Average (exp.)	0.999778	0.010140	1.000196	0.240859	15.10115
ITV (RSD)	0.10%	0.20%	0.10%	0.10%	0.10%
Certified ratio	**0.999698**	**0.010140**	**1.000147**	**0.240876**	**15.10122**
Certified uncertainty	**0.1%**	**0.1%**	**0.061%**	**0.12%**	**0.045%**
Rel. Deviation	0.01%	-0.001%	0.005%	-0.007%	-0.0004%
K factor	0.99992	1.00001	0.99995	1.00007	1.00000

Table 2. Results of Acceptance Testing – NML TIMS measurements of OSL loaded filaments

For enhancing the quality control and the independency of IAEA on both TIMS instruments annual data comparison campaigns are organized with NML Seibersdorf. Safeguards samples are selected throughout the year and archived for later comparisons. They are selected randomly and the results from each laboratory are revealed to the other party only after completion of independent measurements and safeguards considerations. Both laboratories handle similar types of samples and use comparable instrumentation and techniques, such as total evaporation, multi-collector TIMS, IDMS procedures and type of spikes.

Recent examples of cross checking activities for TRITON A are presented in Figures 1 and 2 for two different years. A decay correction for the different measurement dates at OSL and in NML was not applied. For those measurements carried out during the year 2009 (Fig. 1) we observe a relative difference of less than 0.03% between the two laboratories for the n(Pu-240)/n(Pu-239) ratio. The 33 samples have an isotopic ratio between 0.12 and 0.5. A slight positive bias in the OSL results appears but well below the ITV related systematic error of the method.

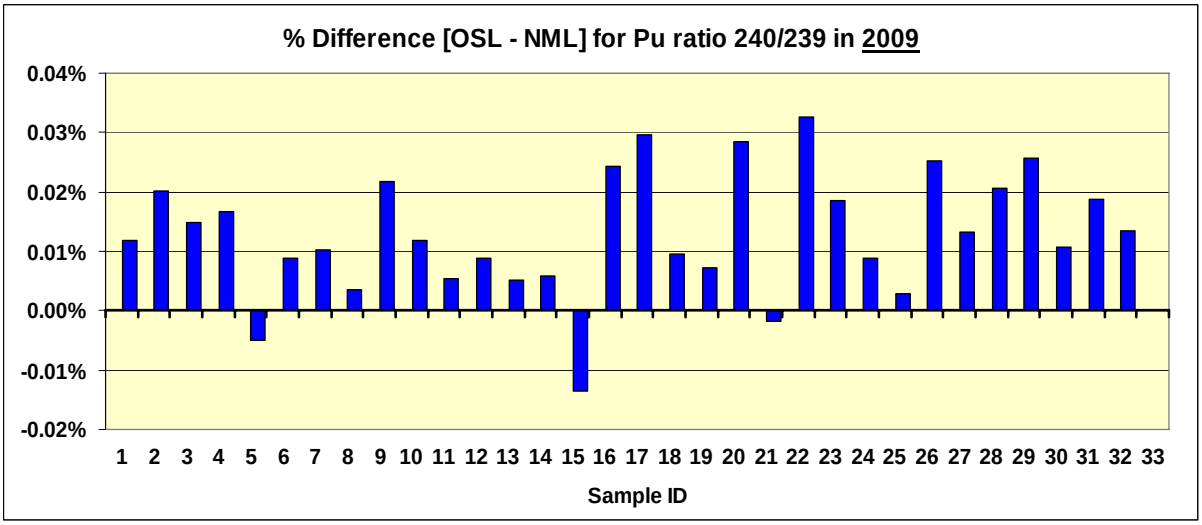

Figure 1. Comparison of measured n(Pu-240)/n(Pu-239) between OSL and NML, 2009

Similar observations are made in the inter-comparison campaign for samples measured by the two laboratories during 2010 (Fig. 2). The relative difference between OSL and NML has decreased significantly in comparison to year 2009 and dropped down to values below 0.02%, with a similar but insignificant positive bias in OSL results compared to NML.

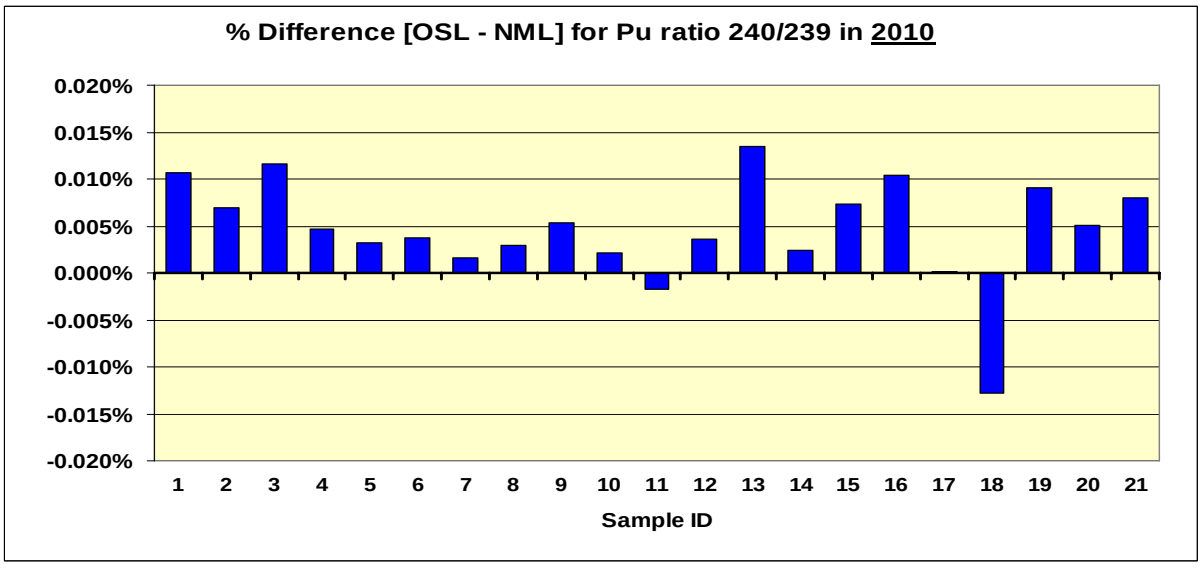

Figure 2. Comparison of measured n(Pu-240)/n(Pu-239) between OSL and NML, 2010

Data for isotopic abundances for minor isotopes are also available and the relative differences OSL against NML for the ratios n(Pu-238)/n(Pu-239), n(Pu-241)/n(Pu-239), and n(Pu-242)/n(Pu-239) are reasonably low. Values below 0.7%, 0.2%, and 0.2% were attained respectively. Taking into consideration the order of magnitude of these ratios (not decay corrected, varying between 0.01 and 0.05 for n(Pu-238)/n(Pu-239), 0.03 and 0.1 for n(Pu-241)/n(Pu-239)and n(Pu-242)/n(Pu-239)) and the associated measurement uncertainties, the performance of TRITON A can be deemed as satisfactory.

ANOVA variance analysis of the data from the 2009 Pu samples measured in both laboratories gave uncertainties as shown in Table 3. The lack of decay correction on the results is the cause of the small positive bias on results in both the 2009 and 2010 Pu results.

Atom ratio of Pu isotopes:	**238/239**	**240/239**	**241/239**	**242/239**
Random component, %	0.4	0.005	0.01	0.01
Systematic component, %	0.53	0.01	(0.14)*****	0.02
Combined, %	0.54	0.02	(0.14)*****	0.03

Table 3. Data from ANOVA calculations (***** decay-correction not applied for the different measurement dates for up to 11 months)

An additional measure to strengthen the internal measurement quality control implies the direct comparison of the two OSL TRITON instruments against each other. This action is undertaken routinely on a monthly basis or when major changes (replacement of hardware parts, edition of new measurement method) have taken place on one of the mass spectrometers. It is important to detect any instrument drift in due time and to initiate appropriate corrective actions. Stable and comparable performance of the two instruments is necessary to facilitate the QC regime necessary to meet safeguards requirements. Both TIMS are under routine performance evaluation. The NBL 137 standard is repeatedly measured on the two instruments and the obtained n(Pu-240)/n(Pu-239) is compared with the certified ratio. Results from such measurements during January and April 2011 are summarized in Table 4. Each average value was determined by measuring six filaments and relative standard deviations in all cases are below 0.005%, indicating an overall coherent state of the instruments. In addition to that, the diminutive deviations of the experimentally obtained results to the certified value of NBL 137 standard support the conclusion of the absence of significant biases. With an exception of 0.012% for TRITON B in January, all relative differences to the certified ratio are considerably lower than 0.01%.

TRITON A	**TRITON B**	**TRITON A**	**TRITON B**
Pu ratio 240/239 (January 2011)		**Pu ratio 240/239 (April 2011)**	
0.240867	0.240844	0.240846	0.240860
0.240859	0.240839	0.240854	0.240860
0.240866	0.240858	0.240861	0.240862
0.240858	0.240846	0.240864	0.240858
0.240871	0.240848	0.240842	0.240849
0.240848	0.240854	0.240834	0.240857
Mean 0.240858	0.240848	Mean 0.240850	0.240858
STD(%) 0.003799	0.002663	STD(%) 0.004757	0.001841
Mean of A/B	0.240855	Mean of A/B	0.240854
Difference (A-B)	1.35E-05	Difference (A-B)	-7.59E-06
Rel. Difference A/B	0.0056%	Rel. Difference A/B	-0.0032%
Certified value	**0.240876**	**Certified value**	**0.240862**
Rel. Difference Cert/A	0.0061%	Rel. Difference Cert/A	0.0051%
Rel. Difference Cert/B	0.0117%	Rel. Difference Cert/B	0.0020%

Table 4. Results of monthly comparisons of OSL TRITON A and B

4. Quality Control Results

Trend charts of the X-bar type are maintained for the measured major isotope ratios of NBL CRM 137 and CRM 010. These CRM are measured as part of each Pu or U batch, respectively. The statistically derived control limits are calculated according to the NIST handbook, e.g. the warning limits are derived from the decay-corrected grand average +/- 1.77 times the average range, the action limits +/- 2.66 times the average range.

Examples for the results on NBL CRM 137 and CRM U010 are shown in Figure 3 and 4. Outliers have not been removed. In addition, for each analyzed filament, individual QC charts are created automatically to monitor signal intensities for the major isotopes, the filament currents, and the mass fractionation for major and minor isotopes.

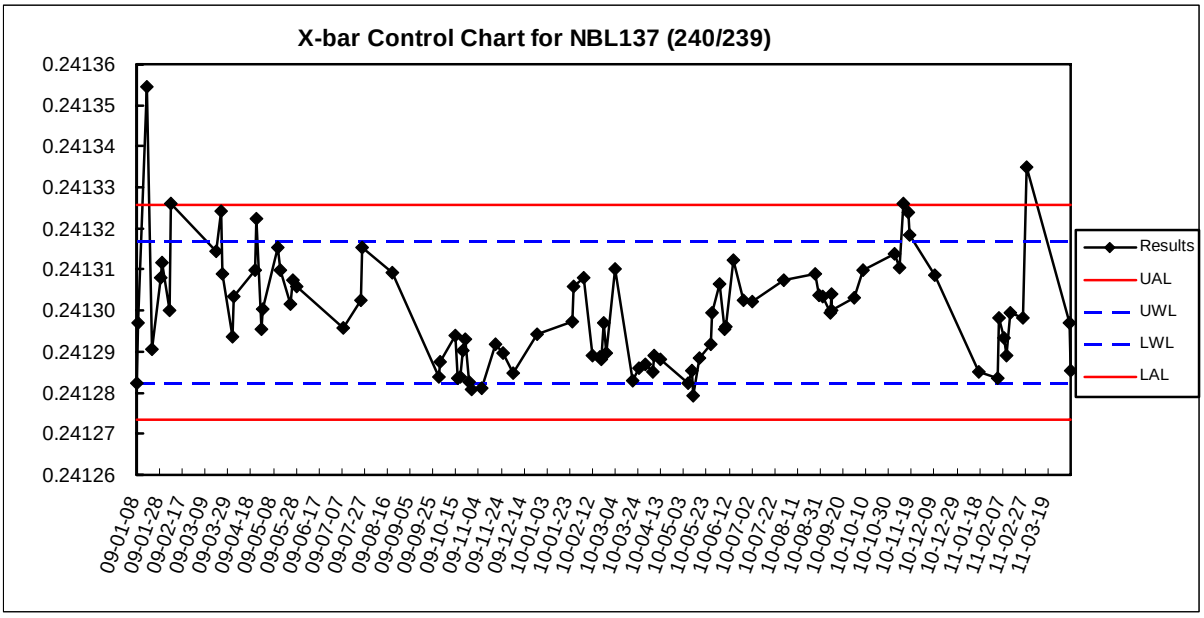

Figure 3. Quality control chart for NBL 137, n(Pu-240)/n(Pu-239). LAL, UAL, LWL, UWL, refer to lower and upper alarm limits, and lower and upper warning limits.

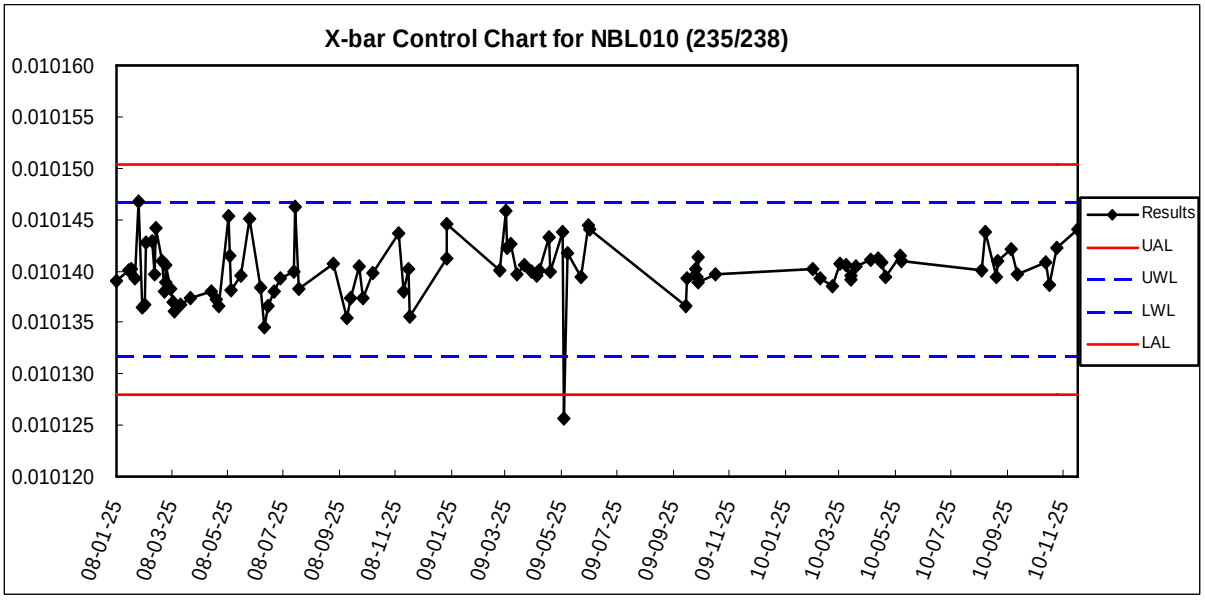

Figure 4. Quality control chart for NBL 010, n(U-235)/n(U-238). LAL, UAL, LWL, UWL, refer to lower and upper alarm limits, and lower and upper warning limits.

5. Conclusions

In order to maintain sustainability and confidence in the overall analytical performance at the OSL, it is essential for both TIMS instruments to undergo regular internal and external inter-comparison exercises and performance checking. Data provided by such undertakings reveal good condition of the instruments and their compliance with international standards of measurement for safeguards purposes including ITV requirements. The second TRITON mass spectrometer recently installed in OSL has passed its acceptance testing phase and is currently in routine operation. It will be employed not only as a quality monitor for the performance of other routine methods at OSL, but will also serve as a back up to the first instrument in case of problems. In addition, with the increase of samples anticipated when the operation of a new MOX facility at the Rokkasho-Mura site (J-MOX), the second TIMS at OSL will be fully dedicated to the increased sample load.

6. Acknowledgement

Part of this work was conducted using Safeguards subsidy granted from the Office of Nuclear Non-Proliferation and Safeguards (JSGO), Ministry of Education, Culture, Sports, Science and Technology (MEXT), as one of the designated works of the Nuclear Material Control Center (NMCC).

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Verification of the reliability of the certified isotope reference materials prepared for nuclear safeguards

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ABSTRACT

A set of isotope reference materials in the form of uranium hexafluoride ranging from 0.5 to 20.0 % of ^{235}U in mass was prepared and made officially available to Brazilian nuclear analytical laboratories.

The use of these materials in the measurement process of blind samples received in the frame of an international interlaboratory comparison program was regarded as an excellent opportunity to independently verify their reliability.

The compliance of the combined uncertainties associated with the $n(^{235}U)/n(^{238}U)$ *isotope amount ratios measured in depleted, natural and low-enriched uranium and the IAEA International Target Values 2010 confirmed the accuracy of the certified isotope amount ratios and therefore demonstrated the reliability of the prepared isotope reference materials.*

Keywords: uranium isotope amount ratios, certified reference materials, mass spectrometry, interlaboratory comparison program, metrology in chemistry

1. INTRODUCTION

Mass spectrometry has long been recognised as the most traditional technique to measure isotope amount ratios. It is able to provide measurement results with high repeatability, just requiring small sample amounts.

There are several mass spectrometry techniques available due to the combination of different ions sources, mass analysers and ion detectors.

These techniques are very precise but this is not enough to guarantee the accuracy of measurement results because is always some kind of bias affecting the measurement process.

Mass discrimination is regarded as the most important factor responsible for this bias. It is corrected by the use of certified isotope reference materials (IRM) under the same instrumental conditions that were used to measure the samples.

According to VIM 2008 3rd edition[1], a certified reference material is defined as a "reference material, accompanied by a documentation, issued by a authoritative body, and providing one or more specified property values with associated uncertainties and traceabilities, using valid procedures".

Certified IRM can also be used to calibrate mass spectrometers, evaluate analytical techniques and methods and set the reference values in interlaboratory comparison (ILC) of measurement results.

Uranium certified IRM have been produced by few top laboratories in the world, the New Brunswick Laboratory (NBL-DOE-USA) (Chicago, USA) [2] and the European Commission Institute for Reference Materials and Measurements (IRMM-JRC-EU) (Geel, Belgium) [3].

Nevertheless, isotope reference materials in the form of UF_6 can only be obtained in from IRMM and with enrichment levels limited to 4.5 % in ^{235}U in mass. Besides, the increasing barriers presently imposed on the transportation of radioactive materials over international borders are complicating the acquisition process of these materials.

These facts led to the establishment of a scientific programme in Brazil focused on the preparation, characterization and certification of uranium isotope reference materials under the modern concepts and practices of metrology in chemical measurement [4].

Uranium hexafluoride (UF_6) was enriched to produce ten base materials with isotope ratios ranging from 0.5 to 20.0 % ^{235}U in mass**.** These materials were then distilled, homogenised and sampled.

The chemical measurements required to assess the purity of these materials were performed in Brazil, at CTMSP laboratories. Volatile impurities in UF_6 were measured by Fourier Transformed Infra-Red Spectrometry (FTIR) and the non-volatile impurities by a quadrupole Inductively Coupled Plasma Mass Spectrometry (ICPMS) using the matrix-matched method [5].

The measurements required to certify the isotope amount ratios of these materials were performed at IRMM laboratories using three independent techniques: Gas Source Mass Spectrometry (GSMS), Thermoionisation Mass Spectrometry (TIMS) and Multi-Collector Inductively Coupled Plasma Mass Spectrometry (MC-ICPMS). The details of this programme is published elsewhere [6,7,8].

2. OBJECTIVES

The objectives of this work are twofold:

(a) to verify the accuracy of the isotope amount ratios of some of the Brazilian IRM produced,
(b) to check the compliance of the uncertainties associated with the $n(^{235}U)/n(^{238}U)$ isotope amount ratio measurements carried out at CTMSP with the requirements of the IAEA International Target Values 2010 for Measurement Uncertainty in Safeguarding Nuclear Materials[9].

The participation in an international interlaboratory comparison of measurement results was deemed as an excellent opportunity to execute these two tasks. This intent was realized in fiscal year 2010, in the framework of the "Safeguards Measurement Evaluation Program (SMEP)" organised by NBL.

3. Experimental

3.1 UF_6 samples

NBL sent to CTMSP eight (8) UF_6 samples with isotope ratios ranging from 0.5 to 4.5 % ^{235}U in mass. The identification and approximate ^{235}U (%) content in mass of these samples are presented in table 1. Half of these samples were analysed in the 2nd quarter and half in the 4th quarter of the year.

SMEP Ampoule	^{235}U (%)
J 68	0.5
J 69	0.5
J 168	1.3
J 169	1.3
J 268	3.0
J 269	3.0
J 371	5.0
J 373	5.0

Table 1 Approximate content of ^{235}U (%) in mass

The received UF_6 samples from NBL were stored in PT-10 Teflon ampoules, sealed with a Teflon disks and metal connections. Firstly they were cooled in liquid nitrogen at -196 ^{0}C and then opened, to allow the installation of metal valves to facilitate the required measurement operations. Then they were kept at a temperature of - 80 ^{0}C in acetone and carbon dioxide bath to allow the light gases such as H_2, N_2, O_2 and especially HF to be pumped off.

3.2 Isotopic measurement

The isotope amount ratios $n(^{234}U)/n(^{238}U)$, $n(^{235}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$ were measured in all samples. Nevertheless, just the results of the $n(^{235}U)/n(^{238}U)$ will be presented in this work.

The mass spectrometer used was the IMU 200, manufactured by IPI Instruments (Bremen, Germany). It is equipped with an electron impact ion source, quadrupole mass analyser, one Faraday detector and one secondary electron multiplier (SEM), device that allows the measurement of low ion signals.

The measurements were carried out using the single standard method. In this method it is very important to select a certified IRM whose isotopic ratio is as close as possible to that of the sample.

It is noteworthy that just the Brazilian IRM were be used in this work.

4. Results and discussion

The results for the isotope amount ratio $n(^{235}U)/n(^{238}U)$ for all samples are presented in table 2. The systematic (u_s) and the random (u_r) components of the uncertainty are presented as well as the value of the combined uncertainty (u_c).

Parameter	J68 & J69	J168 & J169	J268 & J269	J371 & J373
Number of measurements per day	6	6	6	6
Total number of measurements	12	12	12	12
Mean of measured results	0.53727	1.29190	2.98624	4.7959
SD	0.00020	0.00040	0.00061	0.00091
RSD (%)	0.04	0.03	0.02	0.02
Certified value (%)	0.53707	1.2919	2.9843	4.7924
$\mathbf{u_s}$	0.04	0.00	0.07	0.07
$\mathbf{u_r}$	0.006	0.03	0.018	0.026
$\mathbf{u_c}$	0.04	0.03	0.07	0.07

Table 2 Results of $n(^{235}U)/n(^{238}U)$ isotope ratio measurements, certified isotope ratio, systematic, random, and combined uncertainty components.

The data presented in table 2 shows that the repeatability values obtained for all 8 samples, expressed in terms of relative standard deviation (RSD) were in the range of 0.02 to 0.04%. This is a very good result, confirming the high stability ion signal provided by the electron impact ion source of this instrument.

The systematic uncertainties (bias) were in the range of 0.04 to 0.07 %, which is an excellent result for a quadrupole based instrument.

The requirements of the ITV 2010 for several categories of nuclear materials and analytical methods are presented in table 3.

Method	Category	Uncertainty components		
		u_r	u_r	u_c
GSMS	DUF_6 & NUF_6	0.1	0.1	0.14
	$LEUF_6$	0.05	0.05	0.07
TIMS	DU (< 0.3 ^{235}U)	0.5	0.5	0.7
MC ICPMS	U (0.3 %< ^{235}U< 1.0 %)	0.2	0.2	0.28
	LEU (1% < ^{235}U< 20 %)	0.1	0.1	0.14
	HEU (> 20%)	0.05	0.05	0.07

Table 3 Requirements of the IAEA International Target Values 2010 for Measurement Uncertainties for Safeguarding Nuclear Materials

The comparison between the combined uncertainties associated to the $n(^{235}U)/n(^{238}U)$ isotope ratios measured in the set of samples and the requirements of the ITV 2010 is presented in table 4.

	J68 & J69	J168 & J169	J268 & J269	J371 & J373
Category of the material	DU	U	LEU	LEU
ITV 2010 (%)	0.14	0.14	0.07	0.07
Obtained combined uncertainty (%)	0.04	0.03	0.07	0.07

Table 4 Comparison between the requirements of the ITV 2010 and the obtained values for the combined uncertainty for the set of samples of the NBL SMEP 2010

The data presented in table 4 shows that the requirements of ITV 2010 document were met for all three categories of materials: depleted uranium (DU), natural uranium (U) and low enriched uranium (LEU).

This achievement was only possible for two reasons. First, the mass spectrometer used provided results with high repeatability. Second, the Brazilian isotope reference materials have accurate ratios, which allowed a very good correction of the observed isotope amount ratios.

5. Conclusions

The Brazilian isotope reference materials used in this work do have accurate isotope amount ratios as demonstrated by the results of the participation in the NBL Safeguards Measurement Evaluation Program (SMEP) 2010.

The requirements of the IAEA ITV 2010 were met for three categories of nuclear materials: depleted, natural and low-enriched when the Brazilian isotope reference materials were in use.

The Brazilian isotope reference materials are reliable and therefore can be used to measure safeguards samples. This result certainly contributes to build a trustful nuclear safeguards system in South America.

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Considerations on the development of large-sized dried spikes

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Abstract:

The EURATOM treaty stipulates clearly in Article 8 and annex V the need for nuclear isotopic reference material and standards. The Institute for Reference Materials and Measurements (IRMM) is producing these material standards helping the European measurement laboratories in the field of nuclear material control and accountancy to perform measurements of fissile material to the required standards. One main activity of IRMM is the development of uranium and plutonium large-sized dried (LSD) spikes for nuclear safeguards. The IRMM-1027 series of LSD spikes is produced in large batches of approximately 1200 units and contains about 50mg total uranium with an enrichment of ~ 20% ^{235}U and about 1.8 mg of ~ 98% ^{239} Pu per ampoule. These amounts of U and Pu are chosen as such to enable direct measurement of the ^{238}U and ^{240}Pu content in spent fuel solutions. The LSD spikes are covered with a cellulose acetate butyrate (CAB) polymer as a protective stabilizer during storage and transport. The presence of Pu and other ambient factors are causing degradation of this organic layer over a period of several years, limiting the certification validity in time. Optimisation of the current procedure was done at IRMM towards improved organic coating guaranteeing the integrity of the layer for a time span of 24 months. Nevertheless for longer shelf lives research and development for alternative coating approaches are required.

IRMM-1027 LSD spikes require considerable amounts of highly pure uranium and plutonium starting materials. A particular concern is the guaranteed supply of pure ^{239}Pu metal over the next decade and beyond. IRMM proposes therefore a different approach to decrease the amount of ^{239}Pu used for fissile material control measurements when applying isotope dilution mass spectrometry. The IRMM proposal is based on using a 20% enriched uranium isotopic reference material in combination with measurements of plutonium carried out on a diluted sample. By this means the stability problems sometimes encountered in Pu solutions due to formation of colloids or precipitation can be dealt with in a diluted solution. This approach not only reduces the consumption of ^{239}Pu certified reference metal but also facilitates transport of spike material to on-site laboratories and operators.

Keywords: large-sized dried spikes (LSD); verification; safeguards; shelf-life; alternative spikes; CAB coating

1. Introduction

The global Nuclear Non-Proliferation Treaty requires the signatory states to provide detailed accounting records for their fissile materials. The aim of nuclear safeguards is not only the verification of non-diversion of fissile material from its intended and declared peaceful use but also to verify the absence of undeclared nuclear activities in line with the additional protocol in all parts of the nuclear fuel cycle.

Safeguarding nuclear material involves among others the quantitative verification of fissile material by independent measurements. Accurate isotopic analyses are needed in nuclear safeguards especially for plutonium and uranium. Certified Isotopic reference materials with values with small uncertainties traceable to the SI are required for the determination of the uranium and plutonium amount content and isotopic abundance. They provide the basis for a strong and reliable verification and detection system in safeguarding nuclear activities.

The IRMM-1027 series of large-sized dried (LSD) spikes are "primary" isotopic reference materials with certified amount content and isotopic composition for uranium and plutonium. They are used for the determination of uranium and plutonium content in solutions of spent nuclear fuel. They are applied for verification measurements at the on-site laboratories of the European Safeguards' authority at the European reprocessing plants in Sellafield and La Hague and other nuclear installations worldwide. Over the last two decades, LSDs have become a fundamental part of the fissile material control of irradiated nuclear fuel.

The IRMM-1027 series of LSD spikes are prepared by dissolving high purity Pu metal enriched in ^{239}Pu (MP2), natural uranium metal (EC NRM 101) and highly enriched ^{235}U uranium metal (CRM-116) in concentrated nitric acid. The amount content of the spikes is such that no dilution of a typical sample of dissolved fuel is needed before measurement by Isotope Dilution Mass Spectrometry (IDMS) using a single LSD spike. LSD spikes are supplied in a standardized "penicillin" type vials to allow end users manipulations inside hot-cells.

Originally, LSD spikes were prepared in limited numbers for the European Safeguards on-site laboratories and were meant to be used within a short period of approximately a few months. However with time, worldwide demands on the availability of LSD spikes have increased considerably. The spikes are prepared at present in large batches requiring longer shelf-life, and as a result, new developments in the chemical preparation of the spikes have been found necessary.

2. Shelf life of LSD spike

IRMM is the main supplier of nuclear large-sized dried spikes and is, constantly putting efforts and research in further developments of high quality spikes in response to requests from plant operators and safeguards' authorities. Maintaining the integrity of LSD spikes over a longer period is a challenging task as they are in dried nitrate form that has a tendency to flake off the bottom of the glass vial with time. In order to preserve the integrity and sustainability of LSD spikes during transport and storage, a special chemical treatment using a cellulose acetate butyrate (CAB) coating is applied to fix the certified material at the bottom of the vial. CAB provides sufficient mechanical strength, good adherence to glass, and a fair resistance to radiolysis. It also dissolves completely in warm concentrated nitric acid.

Working with plutonium and uranium at the milligram level requires special safety precautions and manipulations are limited when chemical procedures have to be carried out in glove boxes.

Protecting the integrity of the certified nuclear material is one of the biggest tasks.
It is of great importance to protect the integrity of the spike during transport which is a shared responsibility between producer, carrier and end user.
CAB provides the required properties needed to fix the material at the bottom of the vial, stabilize the LSD spikes and subsequently dissolve completely in warm concentrated nitric acid.

Fig 1: Ampoules of IRMM 1027 LSD spikes

The first procedure for CAB coating was sufficient for short term storage, but it was rather problematic to guarantee longer shelf life of LSD spikes. On some batches chipping and material flaking off the bottom of the vial were observed after a period of several months. Since Pu as primary reference material is becoming increasingly scarce and thus expensive, plant operators and safeguards authorities are investigating in the development of domestic spikes as secondary standards directly linked to the IRMM 1027 LSD primary reference material series. This change in emphasis in the use of the spikes has the

consequence that longer shelf lives are required.

Fig 2: Flaking vs. intact CAB coating

IRMM carried out R&D of critical parameters in the process to improve the chemical procedure. Cellulose Acetate Butyrate is a polymer which is capable of resisting degradation from radiolysis, moisture uptake, or by traces of residual acids up to a certain extent, provided that the chemical procedure was successfully carried out. The main contributions leading to a superior CAB layer have been the preconditioning of the glove-box air and changes to the chemical treatment procedure taking the properties of CAB into account. The improved procedure applied from the 1027M batch onwards uses CAB 16%-19% polymer grade. Observations and measurements on the CAB coated LSD spikes show that this type of spike dissolves readily in warm nitric acid and that a homogeneous solution is obtained. It was proven that the presence of CAB does not affect the IDMS analysis process as described in detail in the certification report of the IRMM-1027f batch (1). The spikes can now be certified with a validity of up to 2 years using the present optimised chemical procedure. However, to extend the validity in the future, alternative CAB polymers will have to be investigated.

To produce LSD spikes with longer shelf life IRMM is carrying out long-term stability studies applying different polymerisation degrees of CAB on certified material and studying their behaviour during chemical treatment. It is understood that this new alternative coating also needs to meet the requirement not to result in an impact or modification in the treatment of spikes for customers. Currently, IRMM is investigating two types of CAB, the “35%-39% degree” and the so-called "CAB 551", an industrial cellulose ester with the highest obtainable butyryl substitution. The latter polymer provides better coating adhesion, reduces cratering and mottling and is known to be more resistant towards ultraviolet rays and thus possibly be a better match for dealing with the radiolysis. In order to further study the behaviour of the spike coating a vial from the batch 1027M coated with “CAB 35%-39%” organic layer was used as spike isotopic reference material in recent intercalibration and compatibility measurements of Pu Reference Materials (2).

This was established through several steps which included the oxidation state adjustment (REDOX), separation on anion exchange columns and loading on filaments. All the steps of a chemical treatment were carried out in a glove box.

Throughout the entire process, the behaviour of the new coating was closely monitored and no irregularities were observed. The valency adjustments and the entire radiochemistry were preformed in the same way as in all previous verification measurements to assure the same conditions. All observations indicated that the use of the new organic layer created no hindrance whatsoever.

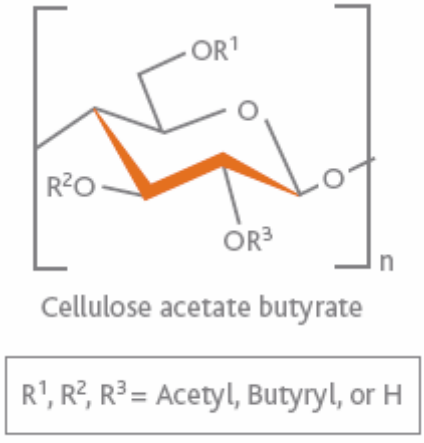

Fig 3: Molecular formula of CAB

A number of vials of the 1027M batch were treated with the CAB 35%-39% polymerisation degree. Even after more than 2 years they do not show any deterioration of the organic layer, whereas cracking and flaking starts to be observed on some of the 1027M vials that were treated with the former CAB coating procedure. Research on the application of specific polymer grades is currently ongoing. Preliminary results are available but it is still too premature to draw final conclusions on the long term shelf life and the feasibility to extend the validity of future 1027 LSD certificates.

Fig 4: Chemical treatment in glove box

3. Alternative spike

To prologue the shelf life of the IRMM 1027 LSD spike series is one field where IRMM puts in a lot of effort. In parallel the fact that the global supply of plutonium metal is limited has triggered to look for alternative LSD spikes. Recently La Commission d'ETAblissement des Méthodes d'Analyse (CETAMA) confirmed that there will not be a new certified European plutonium metal reference material on the market for the next 10 years. But at the same time new enrichment plants, reprocessing plants and MOX fuel fabrication plants are being constructed and reference material suppliers will be asked to develop suitable reference materials for nuclear material accountancy and verification measurements. To satisfy the increased future demands of LSD spikes, domestic spikes need to be prepared from base materials other than metals and new alternative spikes will have to be developed to minimize the use of precious metals, especially Pu MP2.

IRMM proposes a new kind of LSD spike relying on a ^{235}U spike alone with measurements of Pu and U being carried out on the diluted sample. An aliquot of the sample is diluted as required (e.g. 1000x) and the uranium and plutonium isotope amount contents are measured by IDMS using a mixed U/Pu spike.

This method fixes the uranium content in the sample and allows the Pu content to be determined after measuring the U/Pu ratio in the diluted sample.

This 'alternative LSD spike' is based on the fact that the measurement of the effective dilution factor between the spiked solution, in which the uranium and plutonium isotopic ratios are measured, and the original solution of dissolved fuel can be made using the ^{238}U concentration alone.

The advantages of this concept are clear. No large amounts of ^{239}Pu are required and thus the uses of precious and increasingly scarce primary CRMs are reduced. Furthermore equilibration and stability problems caused by the formation of colloids or precipitation of Pu can be dealt with in a dilute solution.

The diluted sample can be multiple spiked and measured to reduce the overall uncertainty if deemed necessary.

Spiking of the dilute solution can be done with a mixed U/Pu spike or by separate spikes. If a mixed spike is used (e.g. 233U/242Pu), no extra measurements are needed compared to the present procedure applying U/Pu LSD spikes.

4. Conclusion

IRMM is striving to tailor the future developments of LSD spikes according to the needs of safeguards authorities, on-site laboratories and nuclear industry by optimising the preparation of the IRMM-1027 series, by proposing alternatives and by carrying out studies on the applicability of these spikes for measurements in future nuclear installations; taking into account the constraints in the availability of base materials and limitation in resources.

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Safeguards thermal ionisation mass spectrometry: State-of-the-practice and recent improvements in nuclear and environmental sample analysis

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Abstract:

One of the challenges in safeguards sample analysis is the determination of uranium and plutonium isotopic and elemental composition with suitable measurement uncertainties while maintaining an adequate sample throughput. For decades, mass spectrometers have been one of the "work horses" reliably meeting this challenge. The state-of-the-practice of thermal ionisation mass spectrometry (TIMS) for safeguards nuclear material samples and environmental samples - from microgram to sub-femtogram - is reviewed, including sample throughput, sensitivity, limits of detection, and measurement uncertainties compared to International Target Values (ITVs). At the IAEA a ThermoScientific Triton TIMS solely dedicated for picogram to sub-femtogram environmental sample analysis using ion counting technology is utilised. For nuclear material samples with analyte amounts of a few micrograms or less, a MAT262 and a Triton TIMS equipped with multi-collector Faraday cups applying (modified) total evaporation protocols is used. Recent improvements in the TIMS capabilities include the implementation of the Guide to the expression of Uncertainty in Measurement (GUM), which addresses the traceability of analytical results and the utilisation of currently available certified reference materials. The net benefit is to strengthen analytical quality control procedures. To utilise the full potential in analysing uranium minor isotopes using the latest generation of TIMS instruments, the modified total evaporation (MTE) protocol was implemented in collaboration with EC-JRC IRMM; it has evolved into a routine analytical procedure resulting in data of superior quality to support the drawing of safeguards conclusions. As an application of the MTE protocol, the agreement between the certified values of two independently produced series of uranium isotope reference materials, the IRMM-18x Series and the NBL U Series, is compared. The exercise provides a crucial insight that fortifies the confidence in the traceability of safeguards analytical results within stated uncertainties using respective certified reference materials.

Keywords: TIMS; safeguards; uranium; plutonium; metrology

1. Introduction

The isotopic signature of uranium and plutonium plays a crucial role in international safeguards and nuclear forensics analysis [1, 2, 3, 4, 5]. In nuclear safeguards analysis, isotopic information is being looked at closely in the verification of a State's declared nuclear materials and activities, and in the detection of undeclared nuclear materials and activities. In nuclear forensics analysis, it is one of the clues helping to attribute unknown nuclear material, for example in a case of nuclear smuggling or in unravelling a nuclear crime scene. It is aided by a tool-box of analytical methods that can reveal other pivotal signatures inherit to samples, including age-dating [7, 8, 9, 10, 11], trace-impurity fingerprints [12, 13, 14], or geolocation information [15, 16, 17]. With respect to nuclear safeguards, the challenge that needs to be addressed is to determine the uranium and plutonium isotope ratios and amount in bulk nuclear material samples with suitable measurement uncertainty, while maintaining a high sample throughput permitting analyses of a large array of samples collected by IAEA inspectors from facilities all over the world. When dealing with bulk environmental samples, on the other hand, the challenge is

to maintain limits of detection for actinide analysis ranging from nanograms of U or Pu per sample to as low as sub-femtograms [1, 5, 6, 18]. In both cases, for nuclear material and for environmental samples, the analytical instruments and methods need to be reliable, robust, and to be used in routine operation. Furthermore, the analytical procedures need to establish traceability of the results to the SI units [19] and be supported by a robust quality control to uphold international quality standards. To meet these challenges, multi-collector thermal ionization mass spectrometry (MC-TIMS) [20, 21, 22, 23, 18] is being used for decades at the IAEA, research institutions, and nuclear facilities all over the world. A comparatively recent addition in the community is inductively coupled plasma mass spectrometry [24, 25, 26, 27, 28], particularly being employed in environmental samples analysis. The state-of-practice and recent improvements in microgram to sub-femtogram analysis of uranium and plutonium is being reviewed with a focus on safeguards thermal ionisation mass spectrometry.

2. Experimental

2.1. Environmental samples analysis

For the analysis of U and Pu isotope ratios in environmental samples, e.g., swipes samples or low-level nuclear samples, a ThermoScientific Triton thermal ionisation mass spectrometer (TIMS) is utilized. For the isotope dilution analysis of U and Pu in environmental samples, a ThermoScientific Element 2 high-resolution single-collector inductively coupled plasma mass spectrometer (ICP-MS) is used, which is not discussed herein. The Triton TIMS is equipped with a multi-collector Faraday cup array, a centre position single electron multiplier (SEM) with energy filter (RPQ) to improve abundance sensitivity, and seven multiple ion counters. Both instruments, Triton and Element 2, are installed in a clean room class 10000 (ISO 6) inside the IAEA's Environmental Sample Laboratory (IAEA-ESL).

Chemically purified uranium or plutonium fractions in nitric acid are drop-loaded with a pipette onto degassed zone-refined rhenium single filaments. Typically, a volume of 1 µL or less is loaded with analyte amounts in the range of 50 pg to 200 pg for Pu and 350 pg to 1000 pg for U. The drops loaded onto the filaments are dried by applying a current of up to about 1.8 A to the filaments. Afterwards, suspended graphite is applied on top of the dried sample and spread out to produce a thin coating and dried by applying a current of 1 A to the filament. The TIMS loading blank for U and Pu is determined and monitored using a suitable uranium-233 and plutonium-242 spike, respectively, and the procedural blank including chemical processing and purification of the samples is monitored by analysing chemistry blank samples. The chemical purification procedure in case of swipe samples is a combination of anion exchange resin MP-1 and UTEVA; details are not discussed herein.

Due to the low U and Pu analyte amount available per measurement, i.e., per filament, the analyses are performed using a SEM peak-jumping method. The multiple ion counters are not used. Analyte concentrations, matrix, and isotopic composition of the samples are matched as closely as possible with certified reference materials (CRMs) used for quality control (QC) and mass fractionation correction (mass discrimination). Samples and CRMs are measured using the same method, which is semi-automatically performed with a defined sequence of heating the filament, mass calibration, ion beam focusing, and sequential analysis of all isotopes of interest. The total evaporation principle is employed, i.e., each filament is measured to exhaustion. The ion beam of the major isotope (e.g., U-238 or Pu-239) is usually in the range of 50000 cps to 300000 cps. An analysis sequence typically consists of a set of samples, some in duplicates, one mass fractionation standard at least in triplicates and one or more QC standards in duplicates.

2.2. Nuclear material samples analysis

2.2.1. Total evaporation method for uranium and plutonium major isotope ratio analysis

For the analysis of U and Pu major isotope ratios (e.g., n(U-235)/n(U-238) and n(Pu-240)/n(Pu-239), respectively) as well as U and Pu isotope dilution measurements in nuclear material samples, e.g., uranium oxides, yellowcakes, ores, and uranium hexafluoride, a ThermoScientific Triton TIMS and a previous generation Finnigan MAT-262 TIMS are used. Both instruments are equipped with a multi-collector Faraday cup array, and the Triton is additionally equipped with two 10^{12} Ohm resistors and a SEM with energy filter (RPQ). The instruments are installed inside the IAEA's Nuclear Material Laboratory (IAEA-NML).

Uranium or plutonium fractions in nitric acid, chemically purified if necessary, are drop-loaded with a pipette onto tungsten double filaments (evaporation filament). Typically, a volume of 1 µL is loaded with analyte amounts in the range of 50 ng to 100 ng for Pu and 250 ng to 500 ng for U. The drops loaded onto the filaments are dried by applying a current of up to about 2.5 A to the filaments. Rhenium double filaments are used for the ionisation filament.

For nuclear material samples, due to sufficient U and Pu analyte amount available per measurement, i.e., per filament, the analyses are performed using the multi-collector Faraday cups and the total evaporation (TE) method. The principle of the TE method is described elsewhere [21, 22, 29, 23]; briefly, the ion signal of the element of interest (all isotopes of interest summed) is stabilized at a target intensity (e.g., 10 V) by keeping the ionisation filament at a constant current but regulating the evaporator filament current. The ion signals of all isotopes are measured until exhaustion (i.e., total evaporation of the analyte). The isotope ratios can then be calculated from the integrated intensities. The drawback of TE is that measurements of background and peak tailing contributions in situ for each sample cannot be performed due to the nature of static measurements (i.e., the magnetic field is not changed). The accurate assessment of these two contributions is particularly crucial for the determination for minor isotopes, here U-234 and U-236, for which the TE method needs to be modified (see 2.2.2.). Analyte concentrations, matrix, and isotopic composition of the samples are matched as closely as possible with certified reference materials (CRMs) used for quality control (QC) and mass fractionation correction (mass discrimination). Samples and CRMs are measured using the same TE method, which is fully automated and often performed overnight. An analysis sequence typically consists of a set of samples, at least in duplicates, one mass fractionation standard and one or more QC standards, usually in multiple replicates.

2.2.2. Modified total evaporation method for uranium major and minor isotope ratio analysis

In the modified total evaporation (MTE) method, the total evaporation analysis (see 2.2.1.) of a sample is interrupted after defined intervals to measure the background and peak-tailing contributions as well as performing a yield calibration of the secondary electron multiplier (SEM) which is used to measure low-abundance isotopes like U-236. The method is described in detail and applied elsewhere [23, 29, 30, 31, 32]; briefly, for the analysis of U isotope ratios n(U-233)/n(U-238) through n(U-236)/n(U-238) in nuclear material samples, e.g., uranium oxides, yellowcakes, ores, and uranium hexafluoride, a ThermoScientific Triton TIMS is used equipped with nine Faraday cups, two 1E12 Ohm resistors, and a SEM with energy filter (RPQ). The instrument is installed inside the IAEA-NML.

The MTE method is configured for a 20 V summed uranium signal and loads per filament of 2500 ng of U. Samples are loaded onto double filaments with Re for the ionization and W for the evaporation filament. The measurement sequence of up to 21 samples is fully automated and typically conducted overnight. Analyte concentrations, matrix, and isotopic composition of the samples are matched as closely as possible with certified reference materials (CRMs) used for quality control (QC) and mass fractionation correction. Samples and CRMs are measured using the same MTE method. An analysis sequence typically consists of a set of four samples in triplicates, five filaments with a mass fractionation standard, and four filaments with a QC standard.

3. Results and discussion

3.1. Environmental samples analysis

3.1.1. Sample matrixes, analyte amounts, and isotopes of interest

Environmental samples analysed are typically swipe samples collected by nuclear safeguards inspectors in and around nuclear facilities all over the world. Typically, the amount of uranium per sample is in the range of nanograms, for Pu it is in the range of picograms or less. Purified Pu and U fractions are analysed with analyte amounts loaded per filament in the range of 50 pg to 200 pg for Pu and 350 pg to 1000 pg for U, respectively. The main isotopes of interest are U-233 through U-238 and Pu-238 through Pu-244.

3.1.2. Sensitivity and detection limits

The sensitivity (overall efficiency, i.e., ions detected per atoms loaded) for the Triton TIMS isotope ratio method is in the range of 0.1 % for U analysis and 0.5 % for Pu. For comparison, the overall efficiency for the Element 2 ICP-MS method for Pu isotope dilution analysis is about 0.5 % using APEX HF desolvation introduction system. The instrumental limit of detection for Pu analysis using the Triton TIMS is below 1 femtogram, for minor isotope ratios n(U-233)/n(U-238), n(U-234)/n(U-238), n(U-236)/n(U-238), n(Pu-241)/n(Pu-239), n(Pu-242)/n(Pu-239), and n(Pu-244)/n(Pu-239) it is typically in the range of a few parts per million, and for the isotope ratio n(Pu-238)/n(Pu-239) typically below 100 parts per million.

3.1.3. Measurement times and sample throughput

The measurement time per filament is in the range of 1 h to 2 h, for U as well as for Pu. A total of about 300 filaments were analysed during year 2010, including samples and standards.

3.1.4. Reference materials, quality control charts, and measurement precision

For the measurement of depleted, natural, and low enriched samples, the certified reference materials NBL CRM U005-A and CRM 112-A as well as IRMM-184 through IRMM-187 are utilized; for high-enriched samples NBL CRM U200 and CRM U500. For Pu analysis, NBL CRM 128, CRM 137, and CRM 136 are predominantly used.

For quality control purpose, the performance is monitored on a day to day basis by tracking results of standard measurements using QC charts. Additionally, critical parameters like SEM yield and dark noise, vacuum readings, and mass calibration are tracked. Results of isotope ratio measurements are checked against certified values and against control and warning limits as well as for signal spikes and summed total signal intensity (overall efficiency). Pu analyses are decay corrected. An example for a QC chart is illustrated in Figure 1; QC data (black squares) are plotted as measured without correction of mass fractionation, peak tailing, or background contribution. The centre lines signify the certified value and its uncertainty (k = 2). The outer lines represent the upper and lower control and warning limits. A summary of measurement precision (relative standard deviation) for QC data of selected CRMs is listed in Table 1.

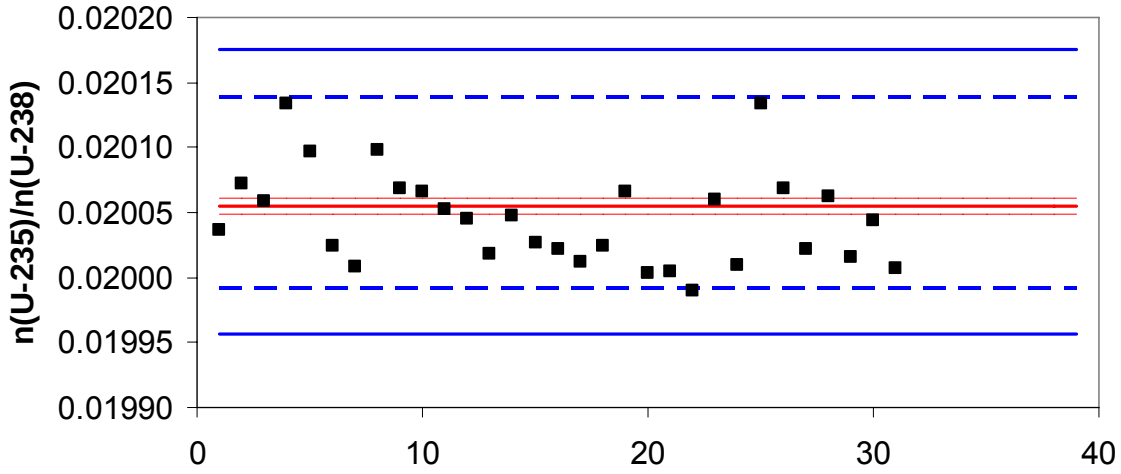

Figure 1: QC chart for isotope ratio n(U-235)/n(U-238) measurement for certified reference material IRMM-185; 350 pg amount of U per filament using Triton TIMS single SEM peak-jumping method. The data set covers a time-period of about one year.

Isotope Ratio	certified value (expanded uncertainty, k = 2)	relative standard deviation	n
NBL CRM U005A; 350 pg of U			
n(U-234)/n(U-238)	0.00003417(70)	2.8 %	20
n(U-235)/n(U-238)	0.0050900(30)	0.18 %	20
n(U-236)/n(U-238)	0.00001186(10)	5.9 %	20
IRMM-184; 350 pg of U			
n(U-234)/n(U-238)	0.000053138(32)	2.4 %	50
n(U-235)/n(U-238)	0.0072623(22)	0.16 %	50
IRMM-185; 350 pg of U			
n(U-234)/n(U-238)	0.000179474(80)	1.0 %	30
n(U-235)/n(U-238)	0.0200552(60)	0.18 %	30
n(U-236)/n(U-238)	0.0000028889(23)	20 %	30
NBL CRM 136 (as of Oct 1st, 1987); 50 pg of Pu			
n(Pu-240)/n(Pu-239)	0.14500(18)	0.08 %	30
n(Pu-242)/n(Pu-239)	0.006801(35)	0.47 %	30
NBL CRM 137 (as of Oct 1st, 1987); 50 pg of Pu			
n(Pu-240)/n(Pu-239)	0.24131(29)	0.07 %	20
n(Pu-242)/n(Pu-239)	0.015599(52)	0.26 %	20

Table 1: Relative standard deviation of QC data (about 1 year) for selected CRMs and selected isotope ratios using the Triton TIMS peak-jumping method; "n" indicates the approximate number of QC data points, i.e., number of filaments measured

3.1.5. Measurement uncertainties and traceability

All reported isotope ratios are mass fractionation corrected against the major isotope ratio of the mass fractionation standards, e.g., IRMM-184, using linear law. Data are also corrected for all significant effects inherent to the TIMS protocol, including TIMS loading blank, peak-tailing, and SEM non-linearity effects. The associated measurement uncertainties are evaluated according to GUM principles [33, 34]. The following sources of uncertainty are assessed: uncertainty of mass fractionation and the certified value of the mass fractionation standard, measurement repeatability (e.g., derived from sample replicates or derived from pooled estimate of standard deviation calculated from adequately matched standards), uncertainties stemming from atomic and molecular interferences, TIMS loading blank, SEM non-linearity and dark noise, and peak tailing. Additional sources of uncertainties might apply in certain cases. The isotope ratio results are therefore traceable to the mass fractionation standard, hence to the SI units.

3.2. Nuclear material samples analysis

3.2.1. Sample matrixes, analyte amounts, and isotopes of interest

Nuclear material samples analysed are typically uranium oxides, yellowcakes, ores, or U hexafluorides collected by IAEA nuclear safeguards inspectors. Purified Pu and U fractions are measured by TE with analyte amounts per filament in the range of 50 ng to 100 ng for Pu and 250 ng to 500 ng for U (and 2500 ng of U for MTE measurement), respectively. The main isotopes of interest are U-233 through U-238 and Pu-238 through Pu-244, with a focus on the major isotope ratios using TE (i.e., n(U-235)/n(U-238) and n(Pu-240)/n(Pu-239)) and the minor and major uranium isotope ratios using MTE. Pu-238 measurements are corroborated by alpha-spectrometry.

3.2.2. Sensitivity and detection limits

The sensitivity (overall efficiency, i.e., ions detected per atoms loaded) for both instruments is in the range of 0.005 % for U analysis and about 0.03 % for Pu. No additive for ionisation enhancement (e.g., carbon) is used because sufficient sample amounts are available and to minimise molecular interferences. The instrumental limit of detection for minor isotope ratio n(U-236)/n(U-238) analysis using MTE is in the range of 3 parts per billion.

3.2.3. Measurement times and sample throughput

The measurement time per filament is about 0.5 h for TE, for U as well as for Pu, and about 1 h for uranium MTE. A sample sequence of up to 21 samples on one sample wheel (Triton) or up to 13 samples (MAT-262) can be analysed within 24 h or 12 h (fully automated), respectively. A total of about 3400 filaments were analysed during year 2010 on the Triton (U and Pu TE and U MTE combined) and about 2100 filaments on the MAT-262 (U and Pu TE combined), including samples and standards.

3.2.4. Reference materials, quality control charts, and measurement precision

For the measurement of depleted, natural, and low enriched samples, the certified reference materials NBL CRM U005-A, CRM 112-A, CRM U030-A, and CRM U100 as well as IRMM-184 through IRMM-187 are utilized; for high-enriched samples NBL CRM U200, CRM U500, and CRM U930 are used. To monitor U-236 measurement performance, IRMM-075(1-5) Series is employed. For Pu analysis, NBL CRM 128, CRM 136, CRM 137, CRM 138, and CRM 144 are predominantly used.

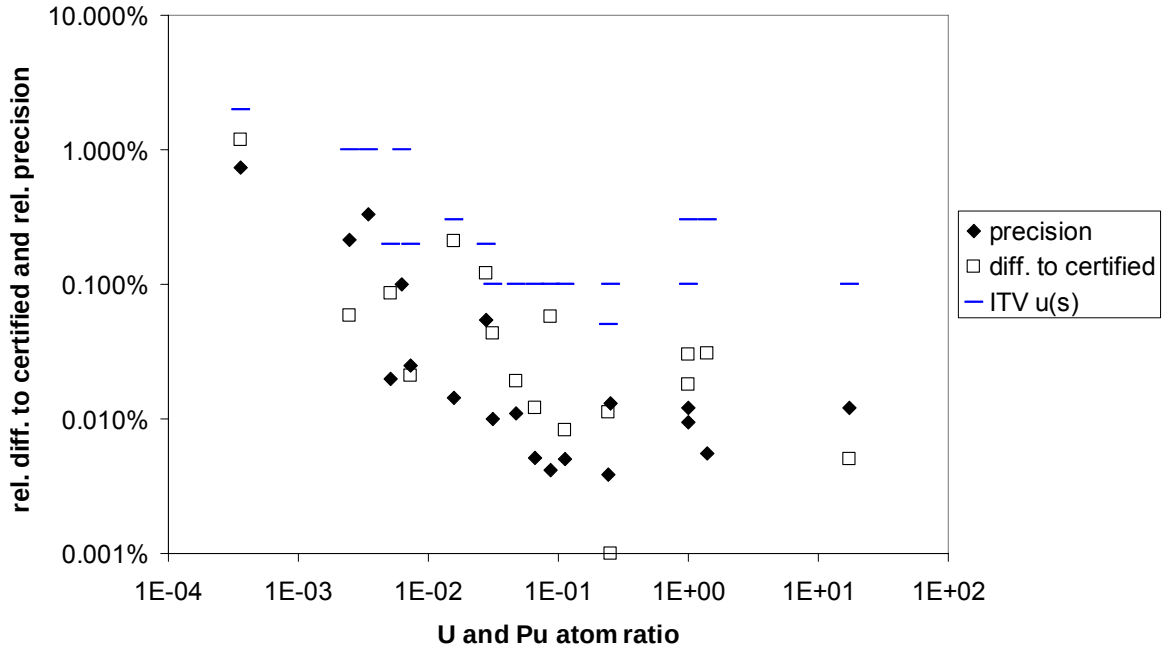

Figure 2: Precision (relative standard deviation) and relative difference to certified for QC data of isotope ratios of various U and Pu certified reference materials. The ITV u(s) limits, i.e., maximum allowed difference, are plotted as well (horizontal bars)

Isotope Ratio	certified value (expanded uncertainty, k = 2)	relative standard deviation	relative difference to certified	n
IRMM-184; 2500 ng of U				
n(U-234)/n(U-238)	0.000053138(32)	0.26 %	0.10 %	350
n(U-236)/n(U-238)	0.00000012446(10)	1.4 %	0.41 %	350
IRMM-187; 2500 ng of U				
n(U-234)/n(U-238)	0.00038700(16)	0.036	0.021 %	70
n(U-235)/n(U-238)	0.047325(14)	0.013%	0.011	70
n(U-236)/n(U-238)	0.000071965(39)	0.20 %	0.20%	70
IRMM-075(5); 2500 ng of U				
n(U-236)/n(U-238)	0.0000000106519(75)	10 %	2.3 %	130

Table 2: Relative standard deviation and relative difference to certified of QC data (1 year or more) for selected CRMs and selected isotope ratios using the Triton MTE method; “n” indicates the approximate number of QC data points, i.e., number of filaments measured.

For quality control purpose, the performance is monitored on a day to day basis by tracking results of standard measurements using QC charts. Additionally, critical parameters like SEM yield and dark

noise, vacuum readings, Faraday cup baseline and gain, amplifier temperature, and mass calibration are tracked. Results of isotope ratio measurements are checked against certified values and against control and warning limits as well as for signal spikes and summed total signal intensity (overall efficiency). Results of Pu analyses are decay corrected. A summary of measurement precision (relative standard deviation) and relative difference to certified for QC data of selected CRMs is illustrated in Figure 2 for the Triton TE method, and summarised in Table 2 for Triton uranium MTE method.

3.2.5. Comparison between "old" generation and "new" generation multi-collector TIMS

A significant gain in precision of a factor of 3 or better, in some cases more than one order of magnitude, is achieved with the “new” generation of multi-collector TIMS, here Triton, when compared to an “old” generation multi-collector TIMS, here Finnigan MAT-262, using the TE method. Data for the two instruments are compared for the same CRMs and same sample size. This is illustrated in Figure 3 showing the repeated measurement of isotope ratio n(U-235)/n(U-238) of the same aliquot of NBL CRM U005-A on both instruments.

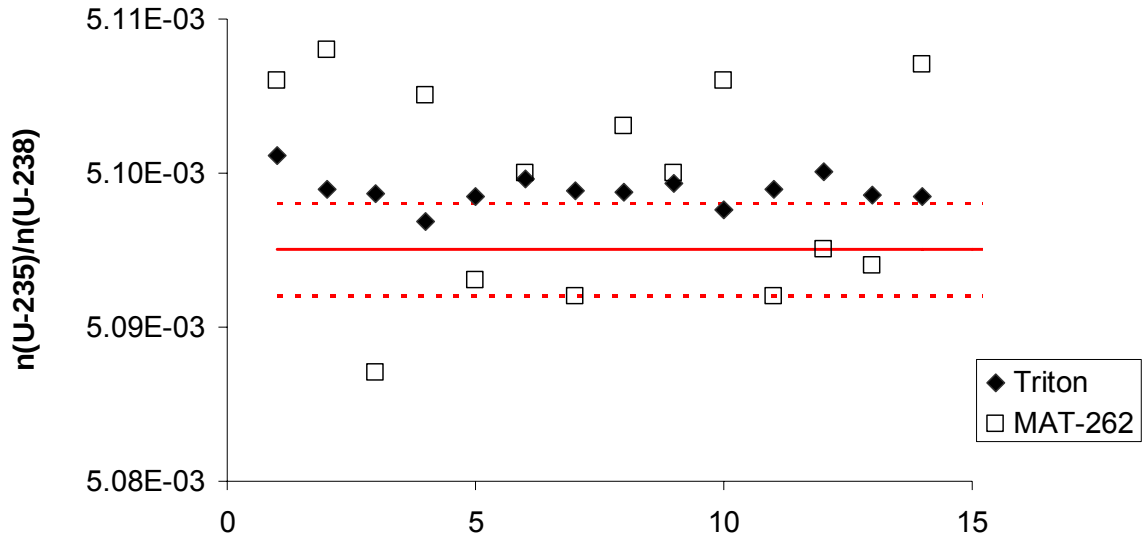

Figure 3: Comparison between “new” generation (Triton) and “old” generation (MAT-262) multi-collector TIMS; QC data for NBL CRM U005-A, about 500 ng of U per filament.

3.2.6. Measurement uncertainties and traceability

All reported MTE results are mass fractionation corrected against the major isotope ratio of the mass fractionation standards, e.g., IRMM-184, using linear law. MTE data are also corrected for all significant effects inherent to the TIMS protocol, including TIMS loading blank and interferences, peak-tailing, SEM yield and non-linearity effects, Faraday cup baseline and gain variability. The reported TE results for the major isotope ratio are compared to the mass fractionation standard as well. Contribution of peak-tailing, Faraday cup baseline and gain variability, and blank and interferences are usually insignificant for the major isotope ratio when compared to International Target Values (ITV) [35]. The associated measurement uncertainties are evaluated according to GUM principles [33]; more detailed information specific to TIMS metrology can be derived from [34]. The following sources of uncertainty are assessed: uncertainty of mass fractionation and the certified value of the mass fractionation standard, measurement repeatability (e.g., derived from sample replicates or derived from pooled estimate of standard deviation calculated from adequately matched standards), uncertainties stemming from atomic and molecular interferences, TIMS loading blank, SEM yield and dark noise, SEM non-linearity, peak tailing, and Faraday cup baseline and gain variability. Additional sources of uncertainties might apply in certain cases. The isotope ratio results are therefore traceable to the mass fractionation standard, hence to the SI units.

3.2.7. A question of traceability - comparison of NBL U series with IRMM-18x U series

The metrological traceability to the SI units is a “property of a measurement result whereby the result can be related to a reference through a documented unbroken chain of calibrations, each contributing to the measurement uncertainty” [19]. In the case of isotope ratio or isotope dilution measurements, the relevant SI units are the unit mol and the unit kilogram. The above mentioned reference to which the results are related to is the certified reference material used to calibrate the mass spectrometer, for example, the CRM used for mass fractionation correction or the spike used for isotope dilution. The CRM itself is supposed to be traceable to the relevant national standard (as stated on the certificate) which itself is supposed to be traceable to the SI unit. Hence, CRMs produced and supplied by different (national) metrological laboratories need to agree within stated uncertainties, otherwise calibrations of analytical instruments performed with respective CRMs may yield results not in agreement with each other. In the case of uranium isotope ratio measurements, the main suppliers for CRMs are the EC JRC Institute for Reference Materials and Measurements (IRMM) and U.S. DOE New Brunswick Laboratory (NBL). Agreement between NBL’s uranium series and IRMM’s uranium series is therefore of interest regarding the statement of traceability for safeguards measurements.

Isotope ratio	relative difference to IRMM-184	Expanded Uncertainty (k = 2, about 95 % CL)
NBL CRM U005-A		
n(U-234)/n(U-238) [&]	0.2 %	2.1 %
n(U-235)/n(U-238) [&]	0.069 %	0.045 %
n(U-236)/n(U-238) [&]	1.28 %	0.90 %
n(U-234)/n(U-238) [$]	0.26 %	0.88 %
n(U-235)/n(U-238) [$]	0.03 %	0.11 %
n(U-236)/n(U-238) [$]	1.8 %	2.0 %
NBL CRM 112-A		
n(U-234)/n(U-238) [&]	-0.01 %	0.21 %
n(U-235)/n(U-238) [&]	-0.002 %	0.064 %
n(U-234)/n(U-238) [$]	-0.03 %	0.033 %
n(U-235)/n(U-238) [$]	-0.011 %	0.036 %
NBL CRM U030-A		
n(U-234)/n(U-238) [&]	0.21 %	0.23 %
n(U-235)/n(U-238) [&]	0.034 %	0.062 %
n(U-236)/n(U-238) [&]	1.27 %	0.86 %
n(U-234)/n(U-238) [$]	0.01 %	0.16 %
n(U-235)/n(U-238) [$]	0.03 %	0.11 %
n(U-236)/n(U-238) [$]	1.8 %	2.5 %
NBL CRM U200		
n(U-234)/n(U-238) [&]	0.12 %	0.25 %
n(U-235)/n(U-238) [&]	-0.01 %	0.10 %
n(U-236)/n(U-238) [&]	-0.06 %	0.32 %
n(U-234)/n(U-238) [$]	0.01 %	0.14 %
n(U-235)/n(U-238) [$]	0.02 %	0.10 %
n(U-236)/n(U-238) [$]	0.00 %	0.16 %
NBL CRM U500		
n(U-234)/n(U-238) [&]	0.03 %	0.19 %
n(U-235)/n(U-238) [&]	0.00 %	0.10 %
n(U-236)/n(U-238) [&]	0.33 %	0.44 %
n(U-234)/n(U-238) [$]	0.01 %	0.14 %
n(U-235)/n(U-238) [$]	0.00 %	0.10 %
n(U-236)/n(U-238) [$]	0.03 %	0.16 %

Table 3: Measured relative differences between certified reference material IRMM-184 and selected NBL U Series CRMs using the Triton MTE method for comparison. The number of replicates per CRM is about n = 10, with n > 50 for NBL CRM 112-A; & versus NBL certificate; $ versus Richter & Goldberg (2003)

For this purpose, the certified reference material IRMM-184 (and in some cases IRMM-187) was measured against various NBL U series CRMs (U005-A, 112-A, U030-A, U200, and U500). IRMM’s -18x Series (IRMM-184 through -187) have been measured against each other by various laboratories

and agree within certified uncertainties with each other, and with other IRMM CRMs like IRMM-3636. The MTE method was utilized for this purpose. Additionally, the TE method was successfully applied to corroborate the MTE major isotope ratio results; details are not discussed herein. The measured relative differences between IRMM-184 and the selected NBL U Series CRMs are listed in Table 3. The contribution of the measurement uncertainties stemming from the MTE method to the expanded uncertainty (k = 2, about 95 % level of confidence) of the relative differences is small, in almost all cases < 10 %; the uncertainties of the certified values of the CRMs are the largest contributors (typically > 90 %). As a consequence, it is a direct comparison of IRMM certified (here IRMM-184) versus NBL U Series certified values within certificate-stated uncertainties.

With the exception of NBL CRM U005-A, an agreement within stated uncertainties between IRMM certified (here IRMM-184) versus NBL certified can be confirmed. For NBL CRM U005-A, the differences on the n(U-235)/n(U-238) and n(U-236)/n(U-238) isotope ratios do not agree within respective uncertainties with NBL certified values, whereas the n(U-234)/n(U-238) ratio does agree. All results agree when compared to Richter & Goldberg [23] published values for the NBL U Series CRMs instead of NBL certified, including CRM U005-A, with the relative difference and uncertainties on minor isotope ratios significantly smaller compared to NBL certified. Note that for NBL CRM U030-A only an upper limit is stated on the NBL certificate for ratio n(U-236)/n(U-238); the result agrees with Richter & Goldberg [23] published value. Note further that for NBL CRM 112-A only an upper limit is stated for ratio n(U-236)/n(U-238)on the NBL certificate; the upper limit of < 5E-9 is confirmed here.

4. Conclusion

For the isotope ratio analysis of uranium and plutonium bulk samples, MC-TIMS is still the most precise and accurate technique available. It is therefore still the method of choice for this analytical task in the nuclear community. This is due to minimal molecular and atomic interferences, comparatively stable ion signals when compared to ICP-MS, decade-long experience in the TIMS analytical techniques, and an overall robust and reliable technology. There has been a gain of up to an order of magnitude in precision of actinide isotope ratios measurement with the introduction of the “new” generation of MC-TIMS, for example with the ThermoFisher Triton TIMS, and additionally in accuracy for U minor isotope determination more recently with the MTE method. For environmental samples analysis, on the other hand, where analyte amounts of Pu can be as low as hundreds of femtograms or less, multi-collector ICP-MS technology offers an advantage over MC-TIMS. This is because the sensitivity, hence instrumental detection limits, of ICP-MS can be significantly lower in routine operation compared to TIMS. As a consequence, high-resolution or multi-collector ICP-MS techniques are complementing or even replacing MC-TIMS techniques for low-level analysis.

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07 Nuclear security and Border Monitoring I

Update on the implementation of the EU RN security plan and instruments / technical projects

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Abstract:

On 1 December 2009, the Justice and Home Affairs Council approved the EU CBRN Action Plan. The EU CBRN Action Plan is aimed at strengthening CBRN security in the European Union. Its overall goal is to reduce the threat and damage from CBRN incidents of accidental, natural and intentional origin. There are about 124 actions that MS and EC are willing to implement. In the area of R&D security the European security research forum ESRIF has indicated the need for a European Security Research Agenda ESRIA and the Commission has reacted to the recommendations from the ESRIF report. Within the 7th Research Framework Programme, for the first time a dedicated CBRN security part has been created and funded.
In support to the implementation of EU Strategy against proliferation of weapons of mass destruction and their means of delivery (WMD) adopted by the European Council on 12 December 2003, the Instrument for Stability (IfS) Indicative Programme (IP) 2009-2011 identifies capacity building against CBRN illicit trafficking and threats as a necessary condition for risk mitigation and preparedness relating to CBRN materials or agents. Complementary EU actions are also numerous at the international level such as the EU supports to the International Atomic Energy Agency (IAEA) activities in the areas of nuclear security and verification through Joint Actions.
The activities on combating illicit trafficking of radioactive and nuclear materials and border control are coordinated within the Border Monitoring Working Group (BMWG) created in 2006 and composed of representatives of the European Commission, European Council, IAEA, United States Department of Energy.
In its strategy 2010-2020, the JRC suggested to focus its security activities in the nuclear and in the non-nuclear field accordingly. Nuclear Security is spelled out as an integrated area that is based on the obligations from the EURATOM treaty and addresses amongst others illicit trafficking and non-proliferation of nuclear materials. The Strategy is developed to support EU MS and the EU policy in these areas.

In this paper we will summarise the ongoing EU activities in the area of RN security and present some examples of technical projects where the JRC has an important role in their implementation and follow up. As example we will mention some of the key actions of the EU CBRN action plan such as the testing of technologies used for the detection of RN materials, the establishment of a nuclear security training centre, the set up with MS of a network for assessment and benchmarking of codes used for the modelling of radioactivity dispersion at a small local scale, the work with the IAEA in improving the use by MS of the IAEA illicit trafficking data base. Outside the EU and under the Instrument for Stability, we will underline the implementation of projects related to the border monitoring and nuclear

forensics but more focus would be on the implementation by the EC through the JRC and UNICRI of the CBRN centre of Excellence.

Illicit Trafficking Radiation Detection Assessment Program - ITRAP+10

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Abstract:

The European Commission, Directorate-General Joint Research Centre (JRC) and the US Department of Homeland Security (DHS-DNDO), in cooperation with the International Atomic Energy Agency (IAEA) and the US Department of Energy (DoE), are jointly implementing the Illicit Trafficking Radiation Assessment Program+10 (ITRAP+10).

The experience of the first ITRAP (1996-2000) resumed what was the state of art of portal, pagers and hand held devices for on-site radioisotope detection and identification. The purpose of this exercise was not to compare instruments but mainly to catalogue the individual sensor capabilities and abilities to detect and identify unshielded and shielded sources.

This exercise showed the importance of effort combination and collaboration of experts, users and vendors as the most effective way to move ahead in improving specifications and technology. Access to nuclear material (PERLA) and other radioactive sources is essential and drafting of specifications and test procedures without practical verification is not valid.

The ITRAP+10 project will carry out an evaluation and comparison of the performance of available radiation detection equipment relevant to nuclear security. The results will provide an independent assessment of the available radiation detection equipment on the market which will serve as a reference for regulatory and other Member State authorities to identify equipment and or families of equipment to address their particular needs, and help to ensure common standards at a European level.

The overall scope of this project is to assist Member State organizations in effectively detecting radioactive materials crossing their borders illegally, whether importations, exportations, or shipments in transit by developing recommendations that describe the technical and functional requirements for the selection of border monitoring equipment so that resources are deployed in an efficient way.

In addition, the manufacturer will get recommendations to improve performance, reliability and user-friendliness of the equipment.

Keywords: nuclear security, radiation detection

1. Background

Nuclear security is an issue of high importance on the international agenda; considerable concern over the illicit trafficking of nuclear material began in the early 1990s following a large number of incidents involving the seizure of highly enriched uranium. The IAEA Illicit Trafficking Database (ITDB) shows an important number of cases of illicit trafficking of radioactive sources and nuclear material worldwide in the last decade [1].

A comprehensive nuclear security program is essential to avoid the illicit trafficking and theft of nuclear material which can lead to nuclear proliferation and the possible construction of improvised nuclear or radiological dispersal devices. Such a security program comprehends the massive deployment of radiation detection systems at borders and crucial nodal points. Other elements such as the nuclear safeguards system (EURATOM, NPT) for nuclear material, the HASS directive for high activity sealed sources, other international legislation concerning radioactive material (from radiation protection to physical protection and transport rules), and intelligence information are taken into account to combat the illicit trafficking of nuclear and radioactive material.

States have the responsibility for combating illicit trafficking and inadvertent movements of radioactive materials. There are a number of measures that must be undertaken by States to combat it and they are, generally, shared between the regulatory and law enforcement agencies as part of a State's national arrangements. One of these measures is the subject of the ITRAP+10 project, namely to improve the detection of radioactive materials at borders.

2. Objectives of the ITRAP+10 program

The first Illicit Trafficking Radiation Assessment Program was a project jointly conducted by the IAEA and the Austrian Research Centre Seibersdorf during the years 1997 to 2000 aimed to evaluate the detection performances of the available equipment involved in nuclear security. It involved also components of regulatory and law enforcement strategies and reports on practical experience at border crossing points.

The experience of the first ITRAP resumed what was the state of art of portal, pagers and hand held devices for on-site radioisotope detection and identification. The purpose of this exercise was not to compare instruments but mainly to catalogue the individual sensor capabilities and abilities to detect and identify unshielded and shielded sources.

This exercise showed the importance of effort combination and collaboration of experts, users and vendors as the most effective way to move ahead in improving specifications and technology. Access to nuclear material (PERLA) and other sources is essential and drafting of specifications and test procedures without practical verification is not valid.

The ITRAP+10 (Illicit Trafficking Radiation Assessment Program) project will carry out an evaluation and comparison of the performance of available radiation detection equipment relevant to nuclear security. The results will provide an independent assessment of the available radiation detection equipment on the market which will serve as a reference for regulatory and other Member State authorities to identify equipment and or families of equipment to address their particular needs, and help to ensure common standards at a European level.

The overall scope of this project is to assist Member State organizations in effectively detecting radioactive materials crossing their borders illegally, whether importations, exportations, or shipments in transit by developing recommendations that describe the technical and functional requirements for the selection of border monitoring equipment so that resources are deployed in an efficient way.

In addition, the manufacturer will get recommendations to improve performance, reliability and user-friendliness of the equipment.

The **certification** of border monitoring instruments itself is **NOT** part of the project goals, however the project's results may help the European Commission to start establishing a network of European laboratories able to perform a variety of tests included in the referenced standards.

The Commission has undertaken the **ITRAP+10 project** through an Administrative Arrangement (AA) with the Joint Research Centre (JRC). The purpose of this AA is to provide support to DG HOME in the field of radiological/nuclear security.

Due to the global relevance of this project, a joint program has been initiated with the US Department of Homeland Security - Domestic Nuclear Detection Office (DNDO) and the US Department of Energy (DoE) which will be carried out in some US National Laboratories. The International Atomic Energy Agency has been involved since the beginning in the project.

This international collaboration will allow having a rather complete worldwide overview of the state-of-the-art of the Border Monitoring Equipment. The bases for reaching this goal are the coordination of our efforts in order to setup similar testing conditions and, when possible, to use the same sources, irradiators, informatics tools to collect and analyze the results. Obviously, another measure agreed is facilitate the exchange of experts from the different laboratories in order to monitor that tests are being carried out in a comparable way. Some of the instruments will be used as reference equipment and will be tested in more than one location.

3. Border monitor equipment

Detection and response to criminal or unauthorized acts involving nuclear and other radioactive material requires the application of radiation detection equipment. The current procedures applied for this purpose are based to some extent on the state-of-art of available instrumentation. The procedure scheme is based on a three steps approach [2]:

- **DETECT**: The radiation portal monitor or personal radiation detector **DETECTS** the presence of radiating material.
- **LOCATE**: The hand-held search instrument **LOCATES** the source of the radiation.
- **IDENTIFY**: The nuclide identification instrument **IDENTIFIES** the nuclide emitting the radiation.

The three-step approach is dictated somewhat by the limited performance of available instruments. This approach is required in order to achieve a solution in which both false negatives (non-detection) and false alarms are at an acceptably low level.

Commonly, the applied metrology can be divided into five basic types of instruments:

- **Fixed radiation portal monitors (RPMs)**, which are pass-through type monitors typically consisting of one or two pillars containing gamma radiation detectors and usually neutron detectors, and monitored from a display panel. They can provide alarm capability to indicate the presence of nuclear or radioactive material above a pre-set threshold. Portal monitors are used for personnel, vehicles, packages and other cargo in a variety of venues.
- **Personal radiation detectors (PRDs)**, which are radiation detectors approximately with the size of a telecommunications pager to be worn by front line officers. PRDs can provide a flashing light, tone, vibration or numerical display that corresponds to the level of radiation present.
- **Hand-held gamma and neutron search detectors (GSDs and NSDs)**, which are radiation detectors used to identify the location of radioactive material. GSDs and NSDs provide greater sensitivity than PRDs do.

- **Hand-held radionuclide identification devices (RIDs)**, which are radiation detectors that can analyse the energy spectrum given off by a radionuclide to identify it. They can be used also as survey instruments to locate nuclear and other radioactive material.
- **Mobile and Transportable radiation monitors (MTR),** which are designed to be transported to a location and used for a specific task or for a specified period of time and do not require permanent mounting platforms. Mobile monitors are those systems that are typically in operation on a platform that is in motion.

4. Description of the tests carried out during the project

Tests carried out during this project will focus on the nuclear/radiological performances of the border monitoring instruments and will be performed at the JRC-ITU laboratories sited in Ispra.

Tests will be mainly based on the technical specifications described in the corresponding European and/or International standards[1]. Table 1 lists the different families of equipment to be tested versus their corresponding standards. Dedicated tests protocols have been developed and agreed in collaboration with DNDO personnel in the frame of this project for the eight families of instruments identified.

Table 1 – Family of equipment vs Standards

Family of equipment to be tested	Standards Reference
RPM (Radiation Portal Monitors) for Road Vehicles	IEC 62244 IAEA NSS1 (2006 & Rev.1)
SRPM (Spectrometric Radiation Portal Monitors)	IEC 62484-FDIS IEC 62244 IAEA NSS1 (2006 & Rev.1)
PRD (Personal Radiation Detectors)	IEC 62401-FDIS IAEA NSS1 2006
SPRD (Spectrometric Personal Radiation Detectors)	ANSI N42.48
RID (Radioisotope IDentifier)	IEC 62327 IAEA NSS1 (2006 & Rev.1)
GSD (highly sensitive Gamma Search Detectors)	IEC 62533
NSD (highly sensitive Neutron Search Detectors)	IEC 62534-FDIS
PRS(Portable Radiation Scanners – Backpack type)	ANSI N42.43 IEC 62327 IAEA NSS1 Rev.1

A big effort has been done by JRC and DNDO in order to harmonize the different standards and to come out with a final proposal to be implemented in the different laboratories involved in both sides of the Atlantic.

The experience gained on both the development and the implementation of the tests protocols will be shared with the European nuclear experts involved in the project through the two Calls for Expression of Interest published in March 2010 [3, 4].
The know-how acquired by this considerable number of nuclear experts during their direct involvement in the different scheduled test campaigns of the project will be essential in the suitable future development of an European network of Nuclear Laboratories with demonstrated capabilities of testing Border monitoring equipment against referenced standards.

5. Status of the implementation

The initial phase of the project has been essentially dedicated to the planning of this three years project, looking for having a clear picture of all the different activities to be carried out in order to be able to optimize time a human resources. Special attention has been addressed to the planning of

[1] In the case that no European standards are available, the corresponding ANSI standards have been selected.

parallel tasks to facilitate the execution of this ambitious project in the established time.

The second task carried out during this initial phase has been the elaboration of the Call for Expression of Interest (CEI) to invite companies to take part in the ITRAP+10 project, which has been published in the Official Journal of the 27MS last 25th of March 2010 [5]. This document includes the technical specifications defining each one of the families of instruments to be tested in the frame of this project, as well as all the legal/economical/insurance conditions defining the collaboration agreement contract between the testing laboratory and the Company.

In parallel to these activities, the test procedures for each family of instruments have been established and will be tested during the second half of 2011.

Two new irradiators have been entirely developed and assembled by JRC: one dedicated to gamma sources and the other one to be exclusively used with neutron sources. The innovative design will allow the automated and controlled irradiation of the equipment in a configuration with geometry of 360° around the source. Two identical units to the ones installed in Ispra are going to be shipped to the US locations defined by DNDO to be used during the tests as part of the strategy of establish the basis to be able to compare the results of the test done in the different laboratories taking part of the ITRAP+10 project.

In addition to the already existing laboratories of ITU-Ispra, two new laboratories have been totally refurbished to host the ITRAP+10 tests. One of them will be dedicated to the small size equipment and the other one will be entirely dedicated to the tests of the Radiation Portal Monitors for both types, vehicles and pedestrian.

As fruit of the collaboration DNDO/JRC establish in the frame of the ITRAP+10 program, a team of experts from DNDO have installed and tested their DCS (Data Collection System) at one of the JRC-Ispra laboratories (all the others Ispra laboratories involved in the test campaigns will be able to send the information to the one hosting the server, in order to collect the information in a centralized way). The DCS has been developed under DNDO supervision to be implemented in their GRADER program (The Graduated Rad/Nuc Detector Evaluation and Reporting (GRaDER^{SM}) Program evaluates commercial off-the-shelf (COTS) Rad/Nuc detection equipment against national consensus standards adopted by the Department of Homeland Security and TCSs) and systematically records all the necessary information about the progress and the results of the test protocol, following the excel sheets developed by each one of the tests. The fact of using the same system to collect and analyse the results of the tests will allow a more accurate cross comparison of the marks obtained during the measurement campaign.

6. Participation

Up to now, a total of 95 instruments have applied to be tested under the ITRAP+10. Table 1 shows a general overview of the actual number of instruments that have applied under each family in both locations EU and US laboratories.

Table 1 – Actual number of instruments classified by family.

Location/Family	PRD	SPRD	RID	GSD	NSD	RPM	SRPM	PRS	MTR	Total
EU	5	4	10	2	3	6	4	4	-	38
US	9	3	12	3	2	12	7	5	4	57

In addition, a total of 15 Nuclear Experts belonging to 4 different EU laboratories have expressed their interest to be involved in the different steps of the project.

Note: this numbers could still suffer some variations depending on the final delivery to the JRC of the complete set of mandatory documents required to be officially registered on the program.

7. Results

The outcome of the ITRAP + 10 project will be a final report on the current technical level of commercially available equipment used in nuclear security as described in previous chapters. The final report will help define the general performance capabilities of each equipment class. The performance capabilities may be used to develop requirements for future equipment selection activities; however, no selection of specific instruments will be conducted as part of ITRAP+10. However it is clear that this project cannot provide a comprehensive survey of this fast developing market.

From the associated results, needs for improvements will be identified as necessary, including the user-friendliness for the use by non-specialist operators.

Participants will receive a personalized test report containing a ranking of the instruments tested done parameter by parameter (or test by test) but without specifically identifying each device (“blind report”). Each participant will know only their own “nickname”. Participants will know how the performances of their instruments compare with the others ones taking part in the tests. Some comments about how to improve their instruments will be included in the report. The report will include:

1. The description of the tests performed: test procedure per family of equipment;
2. Detailed individual tests results per model/device and evaluation of their performances
3. Aggregate results per equipment family (only when the number of instruments belonging to this family taking part in ITRAP+10 is higher than two).

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[5] http://ted.europa.eu/udl?uri=TED:NOTICE:86876-2010:TEXT:EN:HTML&src=0

A novel dual mode imager for detecting special nuclear material

N. Mascarenhas, J. Brennan, R. Cooper, M. Gerling, P. Marleau, S. Mrowka

We describe a scalable 32 element segmented dual mode imaging spectrometer. The detector consists of two planes of liquid scintillator cells. We present the principles of operation of the detector and show results from MCNP PoliMi modeling which was used to guide the design. A new method of variable plane spacing has been incorporated. This can be used to improve detection efficiency or reduce angular resolution on demand. We have applied a powerful analysis algorithm called maximum likelihood analysis (MLA). MLA has been used to image two sources simultaneously in the field of view. The detector has been ruggedized and used to conduct a variety of field tests to image fast neutron as well as gamma ray sources at standoff. Selected results from field tests will be presented. Stereo imaging and ranging has been demonstrated. The detector is also a neutron spectrometer and can successfully distinguish spontaneous fission from (alpha, n) sources. We present a measurement of the energy spectrum of an AmBe neutron source. Finally we will describe work to make this instrument a field portable backpack detector. Our instrument could be a useful tool for nuclear security, for treaty verification and for safeguards.

Virtual Reality based Simulator for Dose Rate Calculations in Nuclear Safeguards and Security

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Abstract:

The current on-site training methods in the field of nuclear safeguards and security are nowadays becoming more and more of a problem due to the nature of the radioactive materials used. Virtual Reality (VR) based training through computer simulator applications can complement the current on site methods. VR methods provide key advantages in terms of safety and cost reduction. But in order to provide an effective training, the virtual system has to be able to meet the demands of realism, time and precision required for the experience to be successful. Unfortunately precision and realism on one side and time constraints are opposing features. Therefore a trade-off has to be reached between them which depends on the particular need of the application. This paper describes the development of a virtual reality based prototype application for training purposes which includes a gamma radiation dose rate calculation method specifically created for virtual reality applications. The algorithm prioritises speed over precision, with the aim of meeting the real time restriction that a virtual reality application requires, in order to provide a realistic feedback to the user. But still giving enough precision to guarantee the training task can be achieved.

Keywords: virtual reality; training; dose rate; point kernel;

1. Introduction

The use of Virtual Reality (VR) for training simulators has been successfully applied in many areas of research and industry. In Nuclear Safeguards and Security some prototypes [1,2] have shown that these technologies are applicable and have the potential to become common use tools in this field.

When dealing with Nuclear Safeguards and Security applications one should consider the need for modelling radiation sources as well as radiation detection equipment. This need applies to NDA applications in Safeguards – Non-Destructive Assay dealing with the measurement and characterization of nuclear materials – as well as radiation detection training in Nuclear Security. Applications cover a wide range of radioactive and nuclear sources and can be used in a wide variety of environments. Examples include: Customs stations for border monitoring, urban and non-urban areas for preparing the response teams after accidents, urban areas for the preparation of special events, etc.

This paper focuses on the accurate simulation of radiation detection equipment. The application uses a point kernel method [3] to estimate the dose rate measured by a portable gamma radiation detector, as an improvement to other methods of determining dose rates that might be slower or less accurate. It can be run in real time within a VR simulator application which can be used for Nuclear Security or Nuclear Safety purposes. An example of a prototype application is showed that implements this method.

The fact that the advantages of VR based training (most significantly cost reduction and safety [4]) match the disadvantages of traditional training methods motivates the development of this work.

2. Radiation Transport Modelling

There are two main broad categories of radiation transport modelling methods: (a) Monte Carlo (stochastic) and (b) deterministic. As several authors have noted [5] Monte Carlo codes provide a very accurate estimation in their measurements, but they have the disadvantage of lengthy processing times. This is not compatible with the real time requirement for the VR simulator, which should typically guarantee a minimum refresh rate of 20 frames per second [6] to enable user comfort and natural behaviour.

One should note that portable detectors are not highly accurate instruments – typically 20% accuracy. This enables the use of a less precise and faster method such as the Point Kernel. This alternative method can consider minor estimations taking into account the imprecision of portable devices.

2.1 Offline Dose Map Method

In an earlier version, this application used a pre-computed, Monte Carlo based 3D map of the dose rates. This system provided accurate dose rate calculations for a series of coordinates (X,Y,Z), with the source being the origin. A linear interpolation function estimated the dose for any point within this map taking the closest available points. The whole map was loaded off-line into an internal database, and consulted for each position where the dose rate was requested.

The most obvious limitation with this method is that it does not account for obstacles between the source and the detector. These obstacles can act as shielding materials. In other words, this could be a strong limitation when simulating real operational environments. Further, that method limited the degree of interactivity for a user to exploit different “what-if” scenarios.

To estimate the attenuation due to obstacles, an attenuation factor is assigned to each object in the scene that can act as a shield. The factor depends on the physical properties (material, thickness of the object). The attenuation is then accounted for each case as illustrated in the following figure.

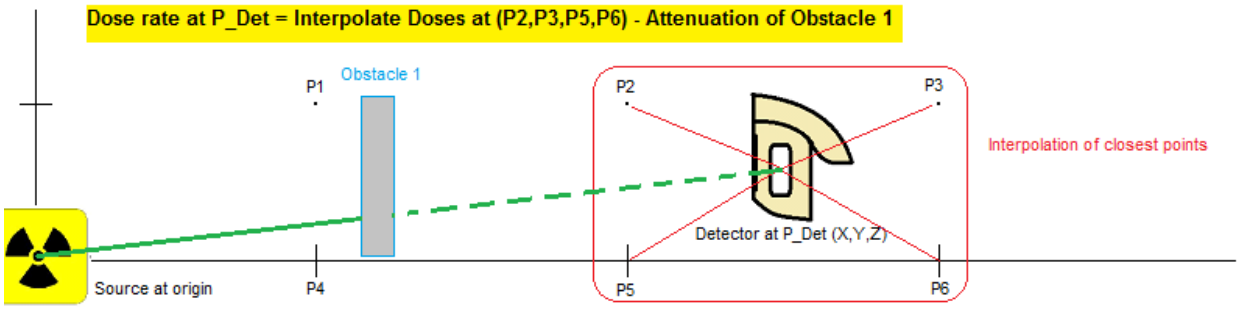

Figure 1 Radiation modelling scheme of the offline dose map method

2.2 Point Kernel Method

The applied algorithm implements a point kernel method. This method is based in two principles; First, calculating the radiation intensity at the point where the detector is placed (I) based on the initial radiation intensity at the source (I_0) and the distance (R) between the source and detector. The following formula specifies this relationship.

$$= \frac{0}{4\pi R^2}$$

Second the attenuation due to possible obstacles (objects in-between the source and detector) is estimated. This depends on the shield's physical and chemical properties, among them the thickness of the shield (d), the density of the material (ρ) and the attenuation coefficient (μ). The following formula relates the Intensity attenuation to the properties of the shield.

$$= {}_0 e^{-\mu d}$$

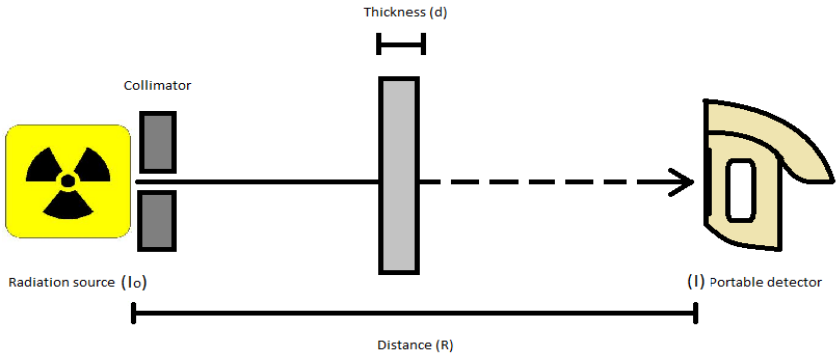

Figure 2 Point Kernel scheme with no Build up factor

The previous expression assumes favourable geometry conditions (a direct line-of-sight, well collimated narrow) as shown in Figure 2, which is unlikely to be found in real cases. In a more realistic scenario with a broader beam or thick obstacles, the above estimation serves only as a lower limit of the dose, due to the fact it does not consider any scattered photons reaching the detector.

In order to compensate for this limitation and obtain a more accurate estimate, a build-up factor (B) is introduced. This parameter is a function of the total attenuation coefficient, the thickness of the obstacle and the energy of the gamma beam. Including this factor in the previous equation the following expression arises.

$$= B \cdot {}_0 e^{-\mu d}$$

The following figure shows the scheme considered in this case with Build Up factor to simulate the more realistic scenario. Build up factors are obtained from the ANSI Gamma-Ray Attenuation Coefficients and Buildup Factors for Engineering Materials report [7].

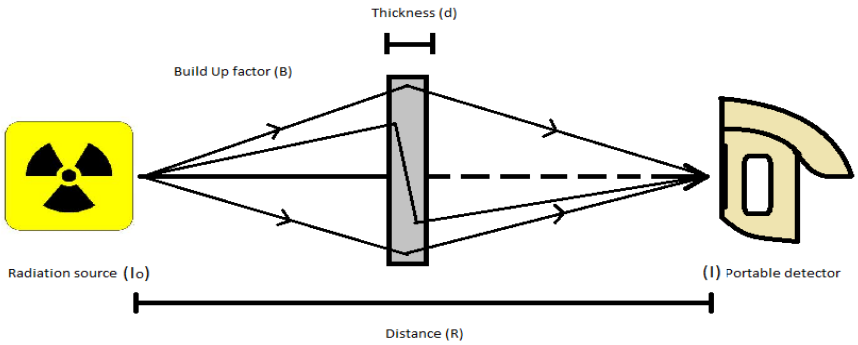

Figure 3 Point Kernel method with Build up factor

Combining the first and the last we obtain the expression to obtain the Intensity (I) at the detector based on the distance (R), source intensity (I_0) and considering attenuation of the shielding (μ,d).

$$= \frac{B \cdot \ {}_0 e^{-\mu d}}{4\pi \mathrm{R}^2}$$

In order to calculate dose rate in tissue, the mass energy absorption coefficient of tissue has to be included in the previous equation, leading to:

$$= \frac{B \cdot \ {}_0 \cdot e^{-\mu d\ shield} \cdot \left(\frac{\mu_l}{\rho}\right)^{tissue}}{4\pi \mathrm{R}^2}$$

The Intensity at the source (I_0) can be defined as the Activity of the source (A) multiplied by the Energy of the gamma emission and its yield (Y) (probability of happening). Therefore I_0 is substituted in the equation, obtaining:

$$= \frac{A \cdot \mathrm{E} \cdot \mathrm{Y} \cdot \mathrm{B} \cdot \ e^{-(\mu d)\ shield} \cdot \left(\frac{\mu_l}{\rho}\right)^{tissue}}{4\pi \mathrm{R}^2}$$

When there is more than one spectral energy line of emission, the individual Energy/probability pairs have to be added, resulting in the final formula which is implemented in the software.

$$= \frac{A \ \sum_{i=0}^{n} \mathrm{E}_i \cdot \mathrm{Y}_i \cdot \mathrm{B}_i \cdot \ e^{-(\mu i \cdot d)\ shield} \cdot \left(\frac{\mu_l}{\rho}\right)_i^{tissue}}{4\pi \mathrm{R}^2}$$

3. Other Elements of the Prototype Application

The radiation model of the application consists of three main elements in addition to the already explained radiation model:

i) the development platform which provides the programming means and 3D engine
ii) the 3D models of the elements of the scenario
iii) the task which the user needs to learn.

3.1 Development Platform

For this work, 3DVIA Virtools was chosen as the development and deployment platform. This programming suite is specifically designed to create 3D Virtual Reality applications. Among other noticeable characteristics, it allows the programmer to import 3D models files from the most extended modelling software tools, and then add pre-programmed complex behaviours to create the application.

To complement the library of pre-programmed functions this platform permits the user to create specific functionality using a C++ based internal language called VSL. Furthermore a SDK (Software Development Kit) is included which allows the programmer to freely, create and even modify the existing behaviours. The platform includes its own 3D engine. There are multiple ways to deliver the applications created, i.e., independent executables or browser based.

All these characteristics made 3DVIA Virtools a suitable platform for the 3D VR based prototype simulator created in this project.

3.2 3D Modelling

A detailed 3D model of the customs area, and the radiation detectors was created, using a popular modelling software (3D Studio Max), which allowed to export the models in a compatible format which can be loaded by the chosen development software.

In order to create an immersive effect, it is of utmost importance that the models are detailed and accurate enough so that the user will instinctively recognise the scenario as his/hers working area and the detectors models match their real counterparts. The following figure shows different virtual models included in the application.

Figure 4 Four snapshots of the virtual model for a border monitoring application involving the training of customs officers in detecting radioactive and nuclear sources

3.3 Training Task

To be successful the training application requires the task to be clearly specified in terms of workflow, decision making process, alternatives to be considered, possibility to provide hints to the trainee, etc. All different actions and the correct order in which they must be completed are the basis of the learning experience.

The figure below shows the flow of possible actions for an application targeted to train customs officers in detecting radioactive sources and nuclear materials for border monitoring applications [2]. The scope is to let customs officers learn a new skill on how to use a simple radiation detection instrument. Each functional block can be further specified in terms of sub-blocks, alternative workflow diagrams, etc.

To be realistic and consistent, the VR application matches the flow of actions done by the user with the real procedures customs officers need to face on a daily basis. Feedback is given to the user by means of on-screen messages, informing about the correctness of the decisions taken..

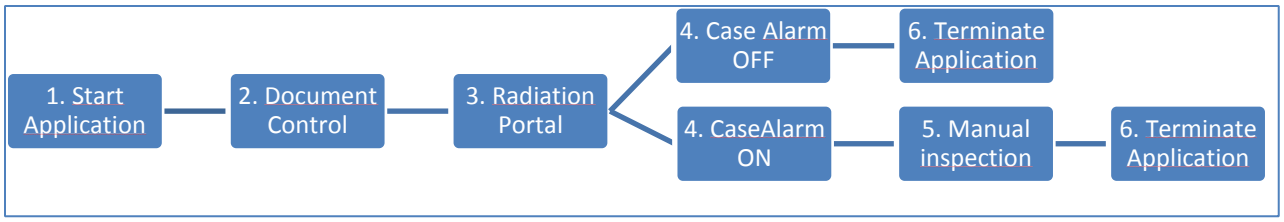

Figure 5 Implemented Task specification

4. Results

An executable Virtools(TM) based application was tested on a workstation computer with an Intel Xeon(TM) processor with a 2.8GHz clock, 2GB of RAM memory and an Nvidia Quadro(TM) FX 1400 graphics card, under MS Windows™ XP operating system.

The application considered a single, point based, radioactive source shielded by a single, material-homogeneous obstacle. It ran with a frame rate well over the 30fps necessary to comply with the real-time restriction.

5. Discussion

This prototype is being developed to prove the feasibility of the concept and the associated implementing technology. The learning efficiency of the system is still to be tested, and a case study with real trainees will be necessary. Nevertheless, this application has been demonstrated to professional instructors, which have expressed great deal of interest and proposed future testing it in real training course environments. Further, there are on-going discussions with both the IAEA and DG Energy training teams on potential use of these concepts and technologies for Safeguards training applications.

When a training application is successful, it can be then modified to create web based refreshing courses. This enables staff to periodically go through some refreshing activities. This is important when the scope of the training involves situations which are not frequent to be met in real life.

The results demonstrate that it is technically possible to use virtual reality based technologies for training purposes in the field of nuclear security, exploiting its advantages in safety and cost reduction. Our conclusion goes very much in line with work executed in other laboratories for applications in homeland security [8,9] or dosimetry assessment in nuclear plants [10].

6. Future Work

Further development of this prototype continues at JRC, some of the innovations and upgrades being undertaken or planned for the future are:

6.1 Nuclide Database

Work is underway to create an online connection that will allow the application to consult a large database of nuclides and shielding materials like the one in [11], therefore providing the user with the information to cover most if not all the possible cases that could arise.

6.2 Radiation Modelling

The next effort in radiation transport modelling is to extend the current algorithms and developments towards 3D free-form volume sources (i.e., not point based), made of non-homogeneous materials.

6.3 Interactivity

Future upgrades plan to increase the number of interactive objects in the scenario, this doesn't affect the radiation measuring but increases the immersive effect of the application due to the similarities of behaviour with real life, which eliminates the need of significant abstraction effort from a non-expert user of the application.

7. Acknowledgements

The authors wish to acknowledge the modelling work contribution by Jerome Lesueur and Damien Brasset and the application discussions with Pascal Daures. We also acknowledge the good discussions with Joseph Magill and Zsolt Soti from the Nucleonica team, JRC-ITU, Karlsruhe, Germany. In preparation for future practical applications, we thank the Nuclear Safeguards training teams of both DG Energy and IAEA for the stimulating discussions and feedback.

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Photoneutron interrogation of shielded/ unshielded HEU by a linac

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Abstract:

A non-destructive assay method was developed for revealing illicit trafficking of nuclear materials. Neutrons being produced in beryllium or heavy water by bremsstrahlung from a 4 MeV linear accelerator induced fission in the samples. Delayed fission neutrons were detected by a neutron collar built up of 12 ^{3}He counters embedded in polyethylene moderator. A PC controlled multiscaler served as a time analyzer, triggering assay startup by the electron beam pulse. Time characteristics of delayed fission neutrons during and after pulsed interrogation were theoretically and experimentally studied. Irradiation-measurement cycles were performed with a 25 Hz pulse repetition rate as optimum setting. High-enriched uranium samples unshielded and shielded by lead up to 14 mm thickness were detected, with a performance practically unaltered. A lower sensitivity limit of the order of 25 mg ^{235}U in a full interrogation time of 1 min (20 s initial irradiation, 40 s irradiation/measurement time during 1000 cycles).can be achieved with beryllium (170 g) or heavy water (105 g) and with a mean electron current of 1.9 μA. Sensitivity can be further enhanced by increasing the measurement time or electron current.

Keywords: illicit trafficking; uranium NDA; Be/D photoneutron converter; delayed neutron counting

1. Introduction

Active methods („interrogation") are widely used for revealing illicit nuclear materials (NM) by irradiation and detection of the induced response. Neutrons can penetrate high-Z shielding material, induce fission in the NM, and fission neutrons can more effectively be detected than passive γ-rays. The active method does not suffer from difficulties due to a shielding encountered in passive γ-spectroscopy This involves an external source that irradiates the material whether or not it is shielded, and the subsequently emitted radiation is detected and used to characterize the material being present. In order to accomplish such an analysis, active radiation portal monitors are to be deployed at customs and border checkpoints. If the induced radiation comprises neutrons, they may penetrate the shielding as well. In such cases it may be advisable not to take the material out of its holder during irradiation and counting. Fission neutrons can be distinguished from irradiating ones via exploiting the time correlation among the former (coincidence counting), or via separating delayed neutrons from the primary and prompt fission ones by their time sequence.

In the fields of safeguards and nuclear forensics, pulsed D-T neutron generators represent a sensitive and versatile variant of active interrogation systems, by counting delayed fission neutrons or gammas (see references in [1,2]). Larger systems designed for inspecting sea cargo relying on D-D [3-5] or photoneutron interrogation by linacs are also common as pulsed neutron sources (references in [1,2]. Assay of hidden or shielded NM via delayed neutrons and gammas from photo- and neutron-induced fissions, by the use of pulsed 10-20 MeV linacs, has also been reported [6-13]. Uranium mass of samples embedded in large concrete packages was assessed by photon interrogation and photofission, using 15-30 MeV linac [14]. Shielded HEU was interrogated with a 60 keV neutron beam produced by a 2 MeV proton linac [15].

A photoneutron interrogation project has been carried out by applying 4 MeV electron linac as a neutron source at the Institute of Isotopes, to induce fission in low- (LEU) [1] and high-enriched uranium (HEU) samples [2]. The electron energy has been converted into bremsstrahlung by a platinum foil, whereas neutrons for interrogation have been produced either in heavy water or beryllium. Delayed neutrons produced in the fissile material have been detected, distinguished from interrogating neutrons by using time discrimination. Having performed the assay of bare uranium previously, results of the interrogation of HEU samples behind lead shielding are reported presently.

Typically about 1% of fission neutrons only are delayed, emitted by fission products. It is necessary therefore to count delayed neutrons for long enough time, in order to achieve good statistics. It means that pulsing neutron sources are necessary, and delayed neutrons can be counted in the interval between pulses. As a result of the continuously pulsing irradiation, the intensity of delayed neutrons goes into saturation with an amplitude depending on the pulse repetition rate. By halving the rate, saturation intensity halves as well, in parallel to the mean intensity of the electron current. It has been shown [2] that 20 s irradiation allows a saturation degree of about 80%, therefore initial irradiations of 20 s at least were applied before starting irradiation-measurement cycles.

2. Experimental

Electron pulses, duration of 2.6 microsec, have been fired with a repetition rate of 25 Hz. Selected from 50, 25, 12.5, or 6.25 Hz, this was established to be the optimum setting in previous experiments [1,2].

Bremsstrahlung was generated on a 20 by 30 mm size, 0.9mm thick platinum converter positioned at 3 cm distance from the exit window of the linac. The diameter of the electron beam was about 2 cm at converter distance.

Neutron production was due to (e,gamma) and (gamma,n) double conversion. Beryllium and heavy water was applied alternatively as photoneutron converter. Their (gamma,n) reaction thresholds are 1.67 and 2.23 MeV, respectively. The neutron energy available from the ^{9}Be(gamma,n)^{8}Be and D(gamma,n)H reaction is up to 2.33 and 1.77 MeV, respectively, at 4 MeV endpoint energy bremsstrahlung. However, the yields abruptly vanish above around 0.9 MeV neutron energy, whereas maximum intensity of the spectrum of evaporated neutrons is at about 0.5 MeV, even at much higher (15-50 MeV) linac energies [16,17].

A home-made neutron collar, as reported earlier [2,18] consisted of concentric polyethylene cylinders of an outer size of 300 mm diameter and 470 mm total length (Fig.1). The innermost ring (of 20mm wall thickness) forms a measurement cavity for the material to be assayed. In between the outermost and inner cylinders, 12 proportional counters (Type SNM-28) of diam. 32 by 308 mm length each, filled with ^{3}He gas to a pressure of 4.0E5 Pa (4 atm) served as neutron detectors. Based on previous experiences, Cd foil was not used. Heavy water or beryllium photoneutron converters of a mass of 105 and 170 g, respectively, were applied at the top of the collar.

The signal processing electronics, consisting of a 400 channel analyzer in multiscaler mode of operation as a time analyzer, was almost the same as reported previously [2]. The multiscaler receives commands from a PC through a micro-controller. Triggering the analyzer was synchronized with the linac control command pulse. A channel width (dwell time) of 100 microsec was selected for the present measurements.

Irradiation-measurement cycles of up to 1000 were carried out at 25 Hz. As experienced previously [1,2], time spectra acquired at 25 Hz show that the pulse length of prompt (interrogating and fission) neutrons cover a half of the 40 ms interval between two pulses. Exponential decay with a time constant of about 2 ms was observed, with a total pulse length of 20 ms. Thus, the effective time remaining for delayed neutron measurement is about 20 ms, no matter if using heavy water or beryllium converter, as observed previously [1,2].

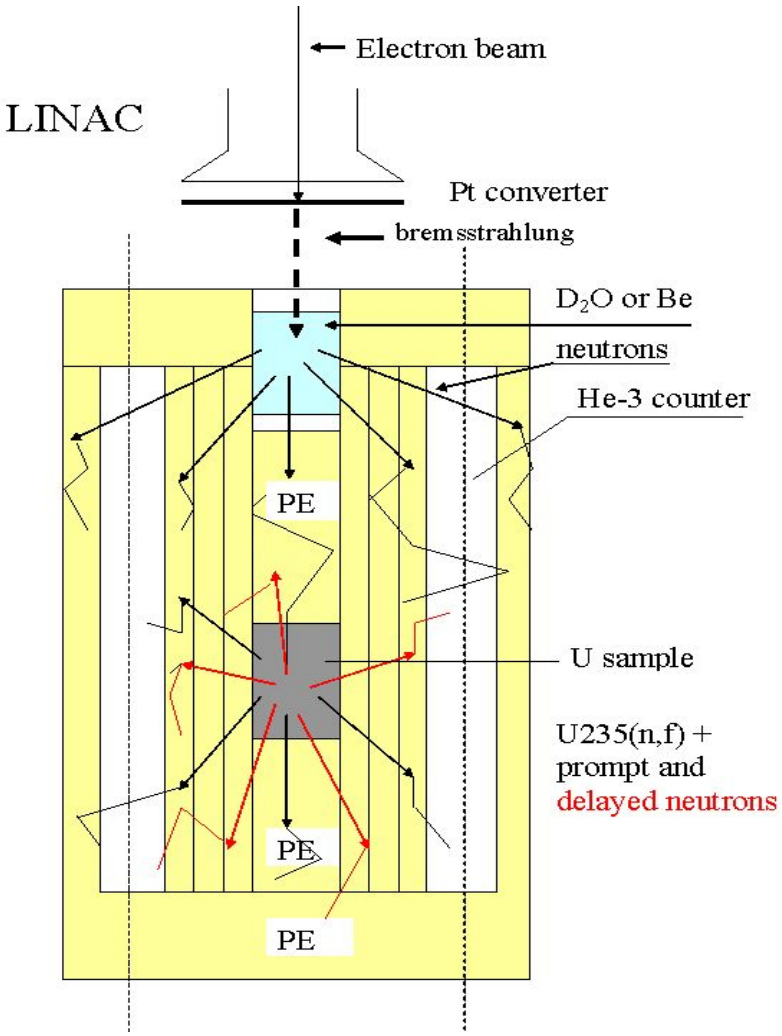

Fig.1. Experimental setup

An irradiation-measurement cycle lasts for 40 ms at 25 Hz. Thus, 1000 cycles last for 40 s. In Fig.2 a time spectrum of the D_2O converter alone is displayed, taken during 1000 cycles at a mean electron current of 1.9 microA, while no U sample was in the measurement cavity. It was practically the same by using the beryllium converter as well.

Time spectra of HEU oxide powder samples of 10.5, 5.5, and 2.3 g mass of 36% enrichment, as well as of a 0.53 g sample of 90% enrichment are seen in Fig. 3. All the spectra were taken by using the D_2O converter**,** during 1000 irradiation-measurement cycles. The mean electron current was uniformly 1.9 microA at 25 Hz. In order to reach a sufficient degree of saturation, 20 s irradiations were uniformly carried out before starting cyclic delayed neutron measurements. The samples were interrogated both unshielded and shielded, being inserted in one of two lead holders of outer diameters 44 and 63 mm by heights 87 and 106 mm, respectively. Holder mass was 0.60 and 2.70 kg, with wall thickness of 4 and 14 mm, respectively. So each diagram contains a time spectrum of the sample unshielded and two spectra of the sample being placed in the lead holders. It can be seen that there is no significant difference between the spectra of the samples being shielded or not, so the actual interrogation of an unknown material can well be performed with the material left in its holder (up to 14 mm lead shielding at least).

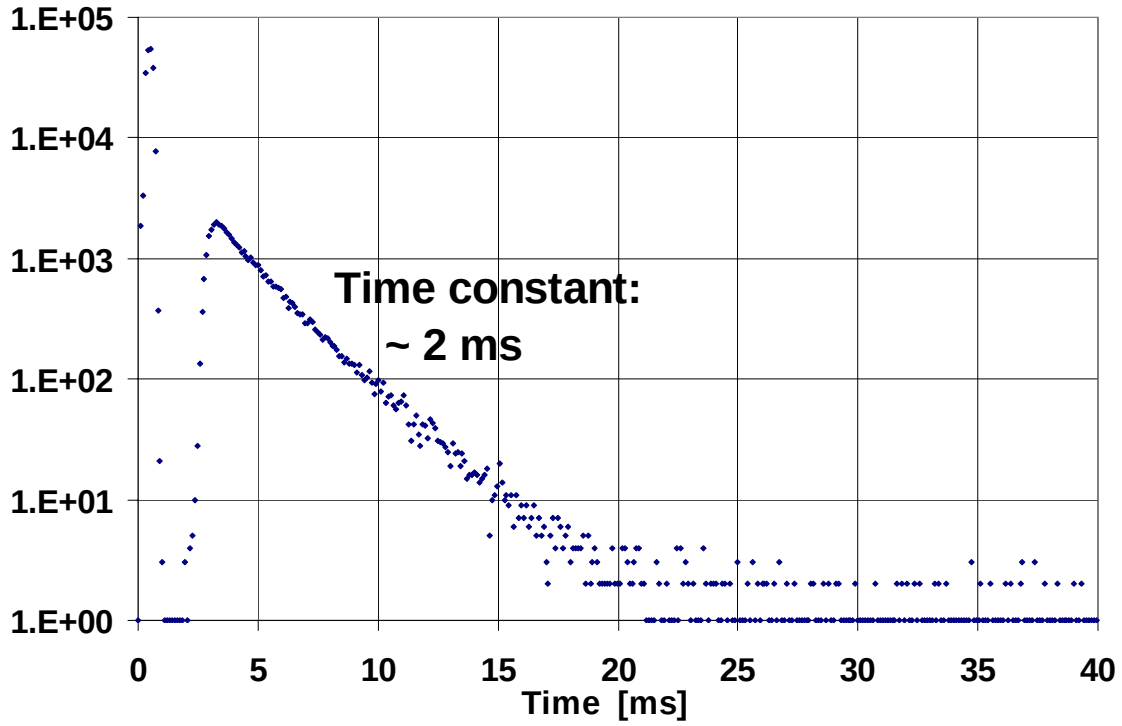

Fig.2. Time spectrum of the D_2O converter alone, without U sample. 25 Hz, 1.9 microA, 1000 cycles

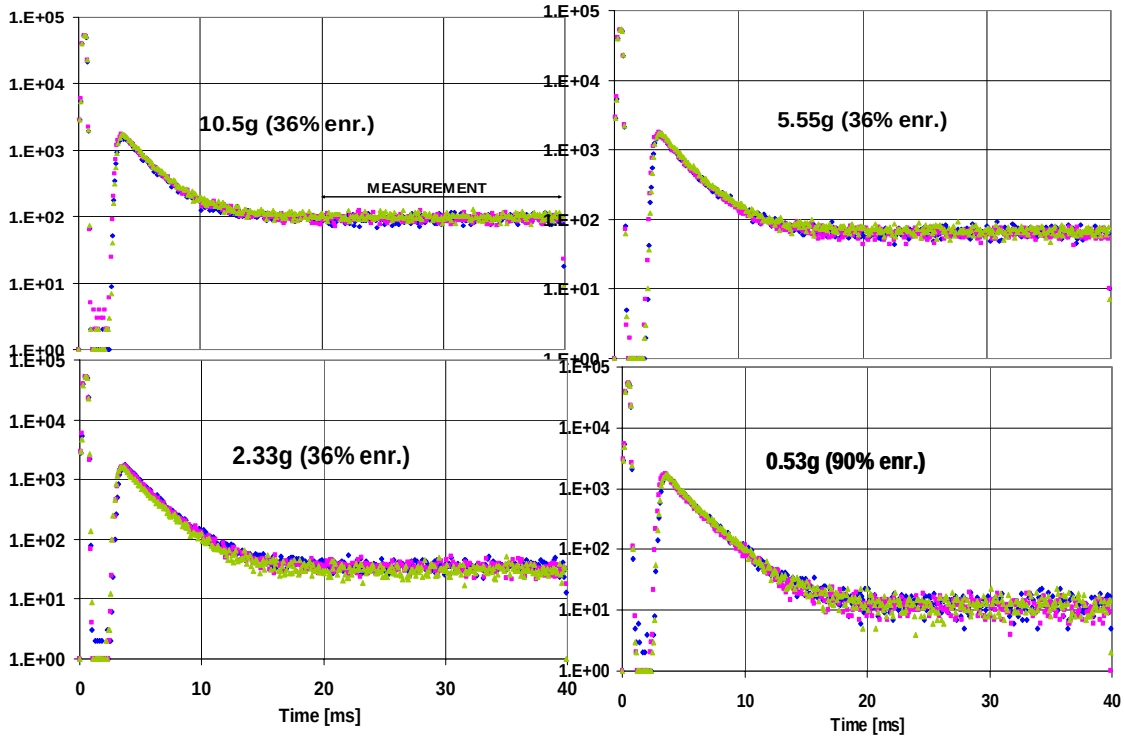

Fig.3. Time spectra of 10.5, 5.5, 2.3g (36% enr.), and 0.53 g UO_2 (90% enr.) samples without and with Pb holders

3. Results and discussion

The number of counts was summed up in the time interval for counting delayed neutrons, i. e. from channel number 200 to 400, corresponding to an effective measurement time of 20 s during 1000 cycles. Total interrogation time thus corresponds to 1 min, including an additional 20 s initial irradiation and 40 s counting time, out of which 20 s is effective. The results obtained by using heavy water converter are seen in Table 1. Indicated are the sample masses, U-235 contents, number of counts, the count rates (per unit effective measurement time), and their ratios for shielded to those for unshielded samples. It is seen that the results of shielded samples agree well with those obtained without shielding. Deviations are within ±10% in the most cases. They can be attributed to the limited reproducibility of the position of the converters and samples, and of the pulse amplitudes, because the linac was started again at every interrogation run (of 1000 cycles). The statistical uncertainty is 1-2% only, negligible.

Sample mass, g	U-235 mass, g	Lead-holder	Counts	Eff. count rate, cps	Ratio shielded/ unshielded
10.49 (Enr. 36 %)	3.20	No	17930	893	
		Small	17979	896	1.003
		Big	19500	972	1.088
5.55 (Enr. 36 %)	1.69	No	12611	628	
		Small	12086	602	0.958
		Big	13300	663	1.055
2.33 (Enr. 36%)	0.71	No	6836	341	
		Small	6423	320	0.940
		Big	5819	290	0.851
0.53 (Enr. 90 %)	0.40	No	2317	115	
		Small	2039	102	0.880
		Big	2251	112	0.972

Table1. Results for unshielded and shielded UO_2 samples by using heavy water converter at a mean electron current of 1.9 microA and 1000 cycles

In Fig.4 the data as a function of ^{235}U mass are plotted, because the samples were not of equal isotopic composition. The signal (number of counts) is not fully linear. This issue was examined in more detail previously for LEU samples of different enrichment [1], and a good linear response was found up to 10g ^{235}U mass. In that case the mass of samples was uniformly 400g, the (low) enrichment of samples was only varied. In the present case, however, sample masses are different. Deviation from the linearity may be due to the flux depression of thermal neutrons in the interior of the bigger (and of enrichment higher than in LEU) samples. The method is sensitive to the ^{238}U content as well, because of the imperfect thermalization and fast fission, although 50-60 times weaker than to ^{235}U [1]. This effect is, therefore, vanishing, compared with the former reason.

Results for the two converters are very similar.

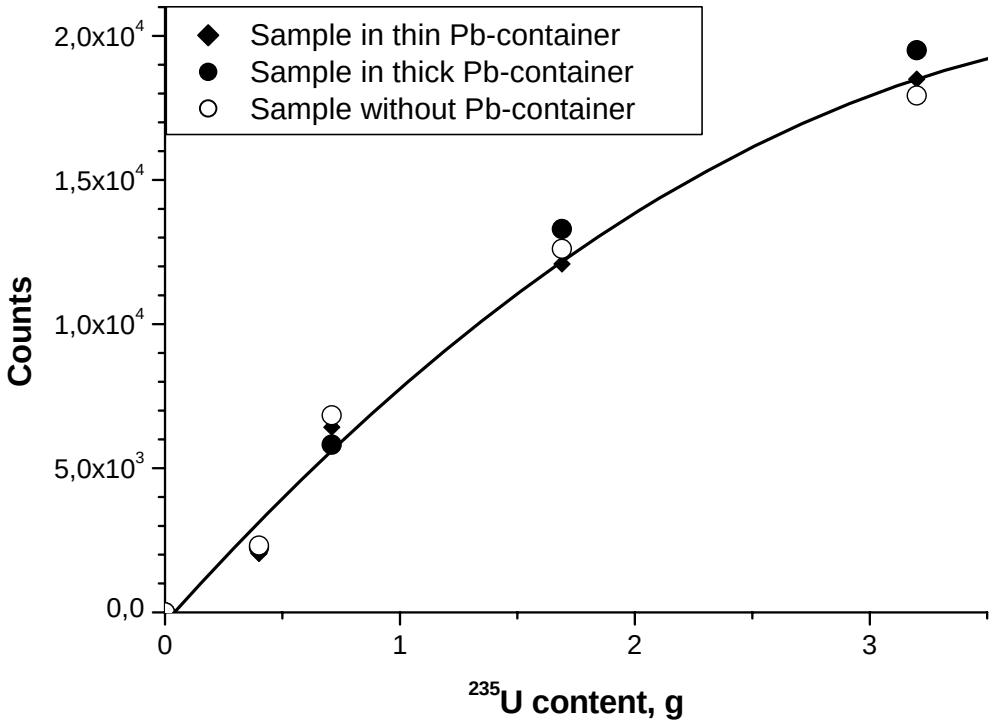

Fig.4. Results without and with Pb holders

Assuming a detection limit of 6.8 count/s (cps) corresponding to twice the background level at 1000 cycles, this means 136 counts during the effective measurement time (20 s). Taking into account a sensitivity of about 5500 counts/g ^{235}U content (Table1, Fig.4), this corresponds to a lower detection limit of about 25 mg ^{235}U.

4. Summary

No influence of a shielding container was observed (up to 14 mm lead thickness at least).

Delayed neutron signal is in a good approximation linearly related to the ^{235}U content. Minor deviation from linearity may be due to the flux depression of thermal neutrons in the interior of bigger samples.

A sensitivity of about 5500 counts/g ^{235}U was achieved. This corresponds to a lower detection limit of 25 mg ^{235}U at a mean electron current intensity of 1.9 microA in a 1 min total interrogation time (1000 cycles). By increasing the electron current or interrogation time (number of cycles), the response can be enhanced.

Half of the 40 ms time interval between pulses can only be exploited, i. e. 20 ms effective time is available for counting at 25 Hz frequency.

Summing up, an efficient laboratory method and equipment have been developed for quantitative assay of unknown U-containing material, without opening its holder. Experience gathered and results may form a basic knowledge for designing a larger system of an active radiation portal monitor.

5. Acknowledgements

Thanks are due to Mr. P. Hargittai for operating the linac and Mr. E. Serf for designing the electronic control system and for his help in the measurements. This work was supported by the Hungarian Atomic Energy Authority.

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08 Implementation of Safeguards

National safeguards licensing system in Hungary

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Abstract:

For keeping nuclear materials and related activities under comprehensive domestic control, an efficient state level safeguards system is required that is based on the continuous application of different regulatory tools. In 2007 Hungary introduced a new safeguards licensing system for establishing and maintaining the effective national safeguards system to account for and control of nuclear materials and nuclear material related activities. The HAEA applies safeguards licensing procedure (including on site inspection in advance) to ensure that the safeguards measures to be implemented by the organization possessing nuclear material are appropriate for complying with the requirements. According to the Ministerial decree 7/2007. (III. 6.) IRM, a first safeguards license issued by the HAEA is obligatory to possess nuclear material and launch any activity related thereto. There are three more types of safeguards license such as safeguards modification license, safeguards transport license and safeguards termination license that is issued for the termination of safeguards requirements subsequent to termination of nuclear activities. There have been 46 first safeguards licenses, 12 modification safeguards licenses and 2 safeguards transport licenses granted by the HAEA since 2007.

Details and experiences of the Hungarian safeguards licensing system will be overviewed and case studies of safeguards licensing will be described in the presentation.

Keywords: domestic safeguards; regulatory control; licensing; state level safeguards system; SSAC.

1. Introduction

The domestic control of nuclear materials and related activities is an important tool to reach several state objectives in Hungary: enable the Sate System of Accounting for and Control (SSAC) of nuclear materials to facilitate the implementation of the international safeguards requirements; (ii) serve as a prevention and detection tool for the physical protection of nuclear materials and (iii) enable the regulatory body to independently verify the compliance with non-proliferation obligations and report the results annually to the Hungarian Parlament.

For this reason the section (1) a) of Article 68 of Act CXVI of 1996 on Atomic Energy gives authorization for the minister supervising the Hungarian Atomic Energy Authority (HAEA) to regulate in a decree the accountancy for and control of nuclear materials in accordance with the international agreements and the domestic objectives. The detailed rules of this regulatory task of the HAEA are provided in the Ministerial decree 7/2007. (III. 6.) IRM and giving competence for the Department of Nuclear and Radioactive Materials of the HAEA (hereinafter referred to as the Safeguards Department) [1].

Fulfilment of obligations related to inspection of nuclear materials as undertaken in international agreements is ensured by keeping the nuclear materials under strict comprehensive control. The comprehensive control is implemented through application of an efficient domestic safeguards system and continuous application of the whole system of regulatory control tools. For establishing and maintaining the effective national safeguards system for the control of and account for nuclear materials and nuclear material related activities, the HAEA applies the following regulatory tools [2]:

- Preliminary registration procedures in the form safeguards licensing to ensure that the facility level safeguards measures to be implemented by the organization for the possession of nuclear material are in compliance with the requirements for the efficient implementation of control activities and the facilitation of inspection objectives;
- Continuous control of nuclear materials and nuclear fuel cycle related activities (including R&D activities) by prescribing information provision obligations.
- Regular evaluation of the activity of the licensee based on processing all of the information submitted by the licensee and independently verified by HAEA via on site inspections.

In the rest of the paper the safeguards licensing scheme of the domestic system is introduced in detail.

2. Safeguards licensing procedures

Safeguards licensing procedures are obligatory to (i) possess nuclear material and launch any activity related thereto (first safeguards license);(ii) to launch any modification important to safeguards (modification safeguards license); (iII) for transport nuclear materials not requiring export-import license according to separate regulation to and from the territory of the Republic of Hungary (safeguards transport license); and (iv) for the termination of safeguards requirements subsequent to termination of nuclear activities (safeguards termination license) [1].

As mentioned above these licensing procedures are preliminary registration procedures that have to be completed before possession of nuclear materials or beginning of related the activities, modifications to the already authorized license, transportation and termination. The licensing procedures shall be initiated at the request of the applicant or licensee. According to other Hungarian laws, submitting an application is an administrative procedure which is liable to duty, has a 30-day administration deadline if there is no notice of completion of documents. In accordance with EU standards, documents can be handed in also in English. After the evaluation and approving of the license application, the safeguards license is granted by the HAEA in form of an authority decision which can be approved or not approved. The Authority ensures consultation during the process if the applicant requires it.

In the following sections the different types of procedures will be introduced in more detail.

2.1. First safeguards licensing

According to the Ministerial decree 7/2007. (III. 6.) IRM, a first safeguards license issued by the HAEA is necessary to possess nuclear material and launch any activity related thereto. The first safeguard licensing application should contain the following information:

- major characteristics of the site (organization, site map, layout, surrounding, access routes, operating conditions, etc.), names and contacts of the owner, operator and of the manager responsible for implementation of safeguards, and activities planned to be performed at the site;
- description of the local accountancy system for nuclear materials, measures guaranteeing the security and safety of accountancy system and accountancy data, and procedure of preparing safety copies;
- proposed material balance areas and those strategic points, which are key measurement points of flow and inventory of nuclear materials;
- measurement, calculation and evaluation methods for determining the quantity of nuclear materials;
- frequency and procedures of accountancy related physical inventory taking;
- technical characteristics ensuring the identification of batches of nuclear materials;
- structure of internal safeguards organization, name(s) and contact(s) of designated facility and site safeguards officer(s), duty order of facility safeguards officers;
- surveillance and containment measures ensuring control of nuclear material flow;
- provisions of physical protection of nuclear materials;
- access procedure of national and international inspectors, with the related health and safety prescriptions.

Application for the first safeguards license shall be submitted to the HAEA at least 3 months, in case of facility at least 7 months before the receipt of the first nuclear material at the site.

In order to help the potential applicants the HAEA developed Guidance on how to determine and in which form to provide the information needed [3]. In addition the authority helps in the determination of very specific information like name of material balance areas, key measurement points, batch names, accounting system, etc. This is found to be very helpful mechanism especially for potential users of small amounts of nuclear materials whom knowledge about safeguards requirements is rather limited.

2.2. Modification safeguards licensing

A safeguards modification license is required to launch any important safeguards relevant modification resulting in the significant change in the following areas:

- local accountancy system for nuclear materials;
- material balance areas and key measurement points;
- flow of nuclear materials;
- measurement, calculation and evaluation methods used for determination of the quantity of nuclear material including development of new measurement, calculation and evaluation techniques;
- frequency and procedures of accountancy related physical inventory taking;
- technical characteristics ensuring the identification of batches of nuclear materials;
- structure of internal safeguards organization, change in facility and site safeguards officer(s);
- surveillance and containment measures ensuring supervision of nuclear material flow;
- exemption of nuclear materials from the requirements of this decree, and de-exemption of exempted materials;
- physical protection system of nuclear materials;
- access procedure of inspectors, duty order of facility safeguards officers.

In the process of making up an application for the modification safeguard licensing, the applicant has to prove that the state after the modification meets the safeguard requirements. Therefore an extensive documentation containing the technical description and specification of (i) the cause and necessity of modification, (ii) the operation of modified part after modification (iii) the safeguard evaluation of modification, the accomplishment of safeguard requirements and purposes (iv) the sequence and process of planned modification (v) the list of documents must be changed because of modification and proposals for modification (vi) regarding references and experiences (vii) the schedule and method of training necessary because of modification (viii) disassemble and maintenance activities necessary for the establishment of modification (ix) programs of tests, try-outs and the evaluation of their successful accomplishment.

If the modification might be related to nuclear safety (applications submitted by nuclear facilities) the applicant is also required to prove that nuclear safety is not reduced by the proposed modification. In these cases, therefore, the modification license is authorized only with the consent the Nuclear Safety Directorate of the HAEA. Alternatively, in case of applications for licensing nuclear safety relevance modification submitted by nuclear facilities to the Nuclear Safety Directorate of the HAEA, which are related to the facility level safeguards, the consent of the Safeguards Department of the HAEA is asked for. With this internal consent mechanism, safeguards, security and safety licensing aspects are taken into account in a single application procedure simultaneously without the need for the licensee to submit several independent applications for the different departments of the HAEA.

2.3. Transport safeguards licensing

For the transport of nuclear materials not requiring export-import license according to separate regulation to and from the territory of the Republic of Hungary a safeguards transport license is necessary. This procedure is needed, because there are some nuclear materials that are not subject to license for transfer between Members of the EU by the export regime of the EU [4]. The aim of the procedure is to provide sufficient information for the authority needed for the control of nuclear

materials leaving and arriving in the country. The licensing can be required for long time period as well if the same batch of nuclear material is transported regularly over the border (e.g. calibration source, depleted uranium container).

In the application for transport safeguard license the applicant shall describe:

- the whole quantity of nuclear material to be shipped, its fissile material content by isotopes, chemical composition, physical form, isotope composition;
- number of objects to be transported;
- type of transport containers (also technical characteristics of sealing possibilities in case of export);
- in case of export the destination country and site, in case of import the material balance area and site where the material is unpacked;
- means of transport;
- in case of export the place of preparation for shipping;
- in case of export the last day when the identification of the material, its quantity and composition can be verified, in case of import the day when the material is to be unpacked, when its quantity and composition can be verified;
- expected time of beginning the shipment and arrival to destination;
- place of transfer of responsibility for the shipment.

At granting the shipment safeguards license the Authority of Nuclear and Radioactive Materials issues shipment or receipt certification for the organization possessing nuclear material, which could be supplied to the border protection and customs organizations if required.

2.4. Termination safeguards licensing

A safeguards termination license is issued for the termination of safeguards requirements subsequent to termination of nuclear activities.

In the application for termination safeguards license the applicant shall describe the way how the nuclear material pass out of the responsibility of the organization, the steps for final termination of the activity, and the documentation related to nuclear materials for the preceding 5 years. If the organization wants to possess nuclear material again after the termination safeguard licensing procedure finished, it has to launch a first safeguard licensing procedure.

2.5. Statistic

Since 2007 there have been 46 first safeguards licenses, 18 modification safeguards licenses, 3 safeguards transport licenses and 1 termination safeguards license granted by the HAEA as shown in Fig. 1.

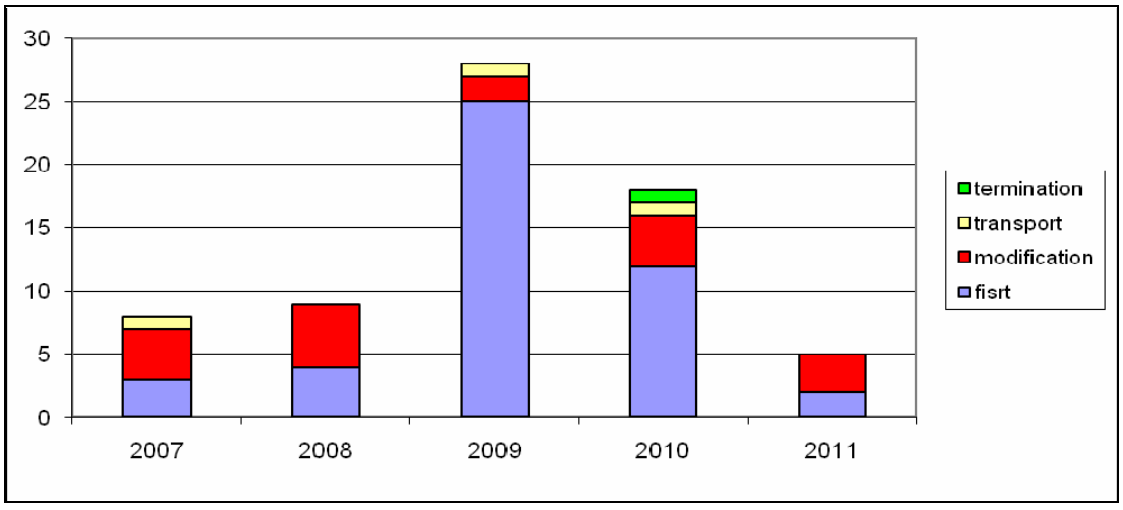

Figure 1. Number of the different types of licensing procedures since 2007

3. Conclusion

The safeguards licensing procedures introduced in this paper form one of the key regulatory tools to ensure that the safeguards measures applied at an organization possessing nuclear material and doing related activity are in compliance with the international and domestic requirements. In addition, they ensure that all information needed for efficient domestic control and to comply with international obligations are collected and continuously updated.

Internal consent mechanism applied by HAEA in the course of license application evaluation has proved to be a very valuable tool ensuring that safeguards, security and safety aspects are taken into account simultaneously.

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SNUICA in action – the Finnish system and experiences in relation to the IAEA Integrated Safeguards inspections

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Abstract:

While the concept of the IAEA Integrated Safeguards has induced changes to the IAEA inspections procedures, it has also had its effects on the actions of the national authorities. The IAEA Integrated Safeguards has been in force in Finland from 15 October 2008, and well before that, a system for the preparedness for the new inspection approaches of the IAEA has been established at the Finnish national authority, STUK.

Due to the requirements of the Finnish legislation, the presence of STUK is mandatory in every safeguards inspection that the international bodies (IAEA as well as the EC) perform in the Finnish facilities. The introduction of short notice random inspections, unannounced inspections as well as complementary accesses has caused new arrangements in STUK operations. It is state's responsibility to enable implementation of the IAEA safeguards. It is state's interest that also EC safeguards requirements are fulfilled, thus it is very important for the national authority to act as an active body to enable the smooth inspection activities of the international organisations. In STUK, the arrangements for successful implementation of Integrated Safeguards inspections are called the SNUICA system. The SNUICA system ensures that STUK's inspector is present in time, has up to date knowledge of the latest safeguards activities, is able to assist IAEA and EC inspectors during the inspection and always concludes inspection by the protocol which all parties sign.

The paper will present the SNUICA system established at STUK for the new safeguards challenges. The paper will also address the experiences from the new type of inspections carried out within the past 2.5 years when the IAEA Integrated Safeguards has been in force in Finland.

Keywords: short notice random inspection; unannounced inspection; complementary access, Integrated Safeguards, State's experience

1. Introduction

The IAEA Integrated Safeguards was introduced in Finland on 15 October 2008. According to the Finnish Nuclear Energy Act, the presence of STUK is mandatory in every safeguards inspection that the international bodies, the IAEA as well the EC, perform at the Finnish facilities. Therefore, starting the Integrated Safeguards in Finland it was needed to make certain arrangements to ensure that STUK could fulfil the requirements of the legislation also in the future.

As a national regulatory body of the control of nuclear materials STUK needed developed its own system to enable efficient IAEA safeguards also in the new era of safeguards. The system is called SNUICA, coming from the definitions Short Notice, Unannounced Inspection and Complementary

Access. As many abbreviations, SNUICA was developed in a good team spirit, and was immediately adopted in STUK, since it is easy to say and remember.

2. About the Integrated Safeguards Approach and general situation in Finland

In Finland, there are two nuclear power plant (NPP) sites, Loviisa 95 km from Helsinki and Olkiluoto 250 from Helsinki. In both sites, there are two operating power reactors and in Olkiluoto, also one under construction. While developing the Integrated Safeguards Approach for Finland, the IAEA has kindly paid attention to the views of STUK and thus the approach is satisfactory also to the national regulatory body. Finland may be a small country, but as Olkiluoto is relatively far from STUK, some special arrangements were considered in the IAEA Integrated Safeguards Approach for Finland.

According to the Approach, there are two new different inspection types that can take place at the Finnish nuclear sites, unannounced inspection (UI) and short notice random inspection (SNRI). At Loviisa NPP both UI and SNRI can take place, while in Olkiluoto only SNRI of these two new inspection types is possible. The UI in Loviisa spent fuel storages is announced to STUK 2 hours before the start of the inspection, whereas the notification time is 24 hours regarding the SNRIs in Loviisa reactors and the whole Olkiluoto nuclear power plant.

3. The arrangements at STUK

For the purposes to the general emergency preparedness of STUK, there are at all times two officers on duty, one for public communications (“duty officer for public information”) and one for emergency preparedness (“expert on duty”). The expert on duty receives all kinds of notifications, alerts and other signals that concern radiation and nuclear related issues in Finland and is specially trained to his/her duties. While preparing for the beginning of the IAEA Integrated Safeguards, it was agreed at STUK that the existing emergency preparedness system with the expert on duty can be used also for safeguards purposes as it was not regarded cost effective to build a completely new system for sole use of safeguard related matters. Using the help of the expert on duty, the notifications of the upcoming IAEA inspections can be easily taken care of, as the person is in any case reachable and equipped to handle these among all the other notifications. Therefore, it was only left to make some arrangements in the Nuclear Material Office regarding the actual participation in the IAEA inspections.

After some internal considerations at STUK, it was agreed that at a time there will be one inspector on call (the SNUICA inspector) at Nuclear Material Office for the IS measures. The duration of each on call shift as a SNUICA inspector was decided to be one week. As it was known that the notifications would be sent during the normal office hours, more precisely between 9 and 9:30 am, the task for being on call did not require any major arrangements and is therefore considered as a normal duty of the safeguards inspectors. The SNUICA inspector is ready and prepared to leave for inspection every week day at 9 am and we have agreed that this readiness does not require the SNUICA to be at the office at that time, since all safeguards inspectors have mobile phones from where they can be reached if a sudden inspection is about to take place at the Finnish NPPs.

4. How the SNUICA system works - experiences so far

STUK has agreed with the IAEA that the notifications regarding UIs and SNRIs as well as complementary accesses (CA) will be sent to the STUK’s emergency fax number. All incoming signals and notifications to that number are directly transmitted to the expert on duty’s mobile phone and therefore, the information regarding the upcoming inspection is received in STUK very efficiently and rapidly. After receiving the IAEA inspection notification, the expert on duty will contact the people at Nuclear Material Office where the preparation to the inspection activities can then begin. Based on the experiences of the actual IS related inspections the people at NM Office are aware of the IAEA inspection notification in minutes after the fax has been received at STUK. The most important thing to do immediately after having the information at NM Office is to call to the IAEA and confirm that STUK has received the inspection notification. The call to the IAEA is made by that inspector who the expert on duty has managed to reach.

As mentioned above, the UI may concern only the Loviisa NPP and more precisely, only the spent fuel storages. When the notification of an UI is received at NM Office the SNUICA inspector needs to leave immediately for Loviisa NPP. If there is no private or company car available at that time, the SNUICA inspector will use taxi as there is no time to lose as he/she needs to be at the gate within 2 hours of the IAEA fax arrival. So far, travelling to Loviisa has not required the use of a taxi. While the SNUICA is on his/her way, the other inspector present at NM Office will check that the proposed IAEA inspector is approved to Finland and informs the Loviisa NPP about the inspection by telephone. The inspector at NM office will also send the STUK's official inspection notification to Loviisa NPP by fax.

The SNRIs may take place in the both Finnish NPP sites, more closely the reactor halls or fresh fuel storages in Loviisa and in any place at the material balance areas in Olkiluoto. The notification time is 24 hours, which leaves enough time for the SNUICA inspector to make the necessary arrangements for the inspection. Therefore it has been agreed that SNUICA him/herself will do the IAEA inspector approval check and make the contacts to the NPP. The time to be at the gate is 24 hours after the fax arrival, however usually at 9 am which is the typical starting time of the IAEA inspections in Finland.

On inspection the IAEA (and EC) inspectors will have an access to the facility on a visitor status. The procedures at UI and SNRI do not differ from those of the usual inspections and at the end, STUK inspector (SNUICA) will prepare, as always, a protocol that will be signed by all parties in that particular inspection. If the EC is not able to participate in the inspection, STUK will always distribute the protocol to the EC and can, if necessary, represent the EC also on the inspection. So far there have been one UI and five SNRIs in Finland since 15 October 2008. From those we have only good experiences.

5 Legal matters

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Some Thoughts on State Level Safeguards

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Abstract:

The implementation of the Additional Protocol gave reasons to rethink the rationale for implementation and evaluation of safeguards. The old system was based on a set of rigid and mostly quantitative criteria, and led to a mechanistic execution of safeguards that seemed not to be effective anymore to cover today's misuse and proliferation scenarios. Especially the maintained approach to relate safeguards mainly to the number of facilities and amounts of materials appeared not to be sufficient. The proposed alternative considers the technical and institutional situation in a state as a whole for the implementation and evaluation of safeguards. This approach is more elegant and sufficiently flexible to cover all existing legal safeguards commitments: INFCIRC 153 alone, INFCIRC 153 with AP, INFCIRC 66 and Voluntary Offer.

The paper deals with State level factors, risk- and information-driven safeguards, and different approaches to cover correctness and completeness of declarations. Considerations based on the physical model and pathway analysis could be helpful to evaluate safeguards performance.

Keywords: State level factors, risk- and information-driven safeguards, physical model, pathway analysis

1. Introduction

IAEA safeguards have a longstanding tradition: They started in the early 1960's with a verification system (INFCIRC/66) related to specific facilities and materials in a country (i.e., not necessarily to all facilities and materials in that country); which is valid for non-NPT states.

With the NPT entering into force in 1970, all non-nuclear-weapon State (NNWS) parties are obliged to conclude comprehensive safeguards agreements (CSA) with the IAEA. Based on INFCIRC/153, CSA's enable the IAEA to verify, that the States are in compliance with their peaceful use commitments made under the NPT.

The system has mainly focused on the correctness of material accountancy in facilities and the fulfilment of specific criteria, while the mandate under CSA's, in principle, also allows for the verification of the completeness of declarations. The latter, however, was not pursued by the IAEA and this profound gap became obvious during the discovery of the NPT violations by Iraq (holding a CSA with the IAEA) in the early 1990's.

The Iraqi case led to the negotiations of the additional protocol (AP) under INFCIRC/540. With these additional measures, i.e., mainly extended access and information, the IAEA has been enabled to obtain credible assurance on the non-diversion of nuclear material and absence of undeclared nuclear material and activities.

2. The Necessity for a New Approach

The development of the INFCIRC/153 model agreement by a safeguards committee in the beginning of the seventies of the last century has to be assessed the context of the political and economic situation of that time. As the nuclear industry was already well established

and steadily growing, the main concern was that industrialized states with a well established industrial nuclear potential could embark in developing nuclear weapons. These states back then formed the main target group for the non-proliferation efforts and for the safeguards measures provided for in INFCIRC/153. Also the fact that INFCIRC/153 focuses on the verification of declared nuclear material is a product of its time. Only very few states would have been willing to allow the IAEA to peer for hidden stocks of nuclear material and clandestine facilities on their territories.

As has been pointed out, the introduction of the AP has caused a dramatic change in the practical implementation and evaluation of safeguards. The IAEA now is aware of the nature and the location of all nuclear and nuclear related activities in a state and has a right of physical access to all relevant locations. The ability to perform a targeted search for undeclared nuclear materials and activities removes the limitations the classical INFCIRC/153 safeguards measures were subject to and allows a complete redesign of the safeguards system.

Since INFCIRC 153 only considers correctness of declarations and material accountancy, a quantitative evaluation and analysis was possible. With the additional objective to detect undeclared materials and activities under INFIRC/540 safeguards face a new challenge in also assuring the completeness of declarations. This broader perspective can only be managed and handled in a qualitative manner, since the term "Credible assurance on the absence of undeclared nuclear materials and activities" cannot be quantified.

By reason of these facts, the IAEA started in the late 90s with the development of a new safeguards concept based on the state-level approach (SLA). The SLA is expected to optimize safeguards efforts especially with the introduction of integrated safeguards in countries with CSA and AP in force.

More than 100 states have ratified the AP and in more than 60 states the agreement is implemented. In these states, he IAEA is now equipped with a robust, modern and sustainable verification system.

With 10 years of experience in applying CSA's with AP's and integrating effectively the old and the new systems, the IAEA is now moving towards state level safeguards. The SLA aims at safeguards implementation and evaluation based on information and risk driven scenarios in individual states. Different state level factors with technical and institutional features can lead to the differentiation of inspection effort in states. In the old system, inspection effort was mainly related to the quantity of nuclear material and facilities in a state. The implementation of state-level integrated safeguards approaches is expected to result in more comprehensive, flexible and effective safeguards.

This demand was also taken over by the NPT Review Conference 2010 which gave the development of the State Level Safeguards approach high importance. The Conference expected also that this development should result in a more comprehensive and flexible Safeguards System.

3. Top Down versus Bottom Up approach

The idea of the old system was to control mainly the inventory and the flow of nuclear material and not the facilities and technologies. The protection of sensitive commercial information had high priority. To assure this concept only nuclear material was to be verified and was made accessible at certain points (Strategic Points, Key Measurement Points). So access was only limited to those strategic points.

The overall control philosophy was implemented in a complicated list of criteria, which gave guidance for the inspection effort in facilities and the field, and a frame for Safeguards evaluation and performance. Since the core of the Safeguards criteria was related to the timely detection of significant quantities of nuclear material in facilities, the attainment of material and timeliness goals were the dominant factors in the yearly IAEA SIR statements.

So in fact states with complex facilities, large fuel cycles and sensitive nuclear materials had major inspection efforts on the one hand but otherwise medium performance results since under real plant conditions not all criteria could be fully met every year. So already in this approach the element of differentiation between states was implemented.

However, as experience has shown, it may not be the technical capability in terms of available

nuclear material and knowledge that constitutes the main driving factor to strive after nuclear weapons. In the current system, the safeguards approach is build bottom up. Generic facility specific safeguards approaches are combined together and this aggregation then modified in some aspects according to state specific considerations. As a consequence, it is still the amount of nuclear material in a state that primarily determines the amount of verification effort spent in a state.

Just to give two examples. Let us take two states that do have neither enrichment nor reprocessing facilities and for which the IAEA was able to draw the broader conclusion that there are no undeclared nuclear materials or activities in that state. In state A the IAEA spends 20 person days of inspection (PIDs) for the verification and in state B 600 PIDs to verify irradiated fuel and other non-direct use material. What is the additional value generated in state B compared to state A? From the current SIR we can learn that the IAEA is applying a cost assignment methodology where its activities are considered as “products”, and the production costs per product are to be determined by applying the cost methodology. What is the product generated for state B that justifies a one thirty times higher production effort compared to state A?

In our second example, let us consider two states with the same number of 400 PIDs a year. Both states carry out enrichment activities; however, state A has implemented integrated safeguards, whereas state B only has a CSA in force, by did not ratify the AP yet. What is the product generated for State A that justifies the same production effort compared to State B?

If we accept that not the amount of available nuclear material is the primary trigger to seek for a nuclear armament, a more flexible system, a top down developed system, has to be created. The main construction principle has to be the flexibility to adapt to new situations. When we review the history of safeguards, we see that the threat scenario has changed considerably over the years. We have to be prepared for further and even more rapid changes. The tasks of the IAEA within the field of verification may be enlarged in future without according increases in the budgets. To be able to cope with such challenges a true target-oriented approach able to adapt to different objectives should be developed.

In contrast to the old system state level safeguards have to start with an analysis of the overall situation in a state which includes the technical nuclear infrastructure and capabilities but also institutional factors concerning the nonproliferation situation.

Since the old system was based on mechanistic and rigid factors, is the new philosophy anchored at flexible and dynamical characteristics of a state. Both inspection effort and safeguards evaluation are related to the situation in that state. This system is information and risk driven and gives guidance for the distribution of budget and inspection effort in states. The other positive consequence is a strong improvement in safeguards effectiveness because the system is now in a better position to focus on specific proliferation scenarios in states of concern and improving efficiency while reducing efforts in other cases.

To understand and evaluate the nonproliferation situation in a state a comprehensive diversion path way analysis can be performed. This bottom up approach can help to understand the technical situation in a state and can contribute in substantial manner to the wished transparency but should not be the basis for distribution of inspection effort.

The availability of modern and robust safeguards instrumentation is essential for the efficient and effective implementation of IAEA safeguards. The improvement of the sensitiveness of accountancy and measurement systems is always a first goal. Remote monitoring of facilities is a second goal. Here, the IAEA or regional or states’ systems will be connected online to nuclear facilities.

For this purpose, digital instrumentation must be available not only for nuclear measurements but also for containment and surveillance. In order to ensure confidentiality, components have to be available with built-in tamper resistance features and data authenticity. For future facilities, such safeguards aspects should be considered already at the design stage when constructing a new facility. Further advantages can be achieved by considering and realising synergies between the different standards for safety, security and safeguards.

Besides the already mentioned improvement of the accountancy system, the ability to detect undeclared nuclear materials and facilities has to be further improved. Environmental monitoring, satellite imagery and nuclear forensics are very promising fields.

The IAEA has access to a wide ranging spectrum of information such as inspection reports or reports from other verification activities, states supplied information, trade analysis, open source information, and intelligence provided by member states. With the collection and analysis of all this information the IAEA has a powerful tool to plan and perform actions or draw conclusions

4. State Level Safeguards in NWS

The further development and evolution of the state level approach would also be of great advantage for implementing safeguards in Nuclear Weapon States (NWS). Already today peaceful activities and facilities in NWS are under IAEA safeguards via voluntary offer agreements, where the IAEA can select from a list of dedicated facilities, which one could be safeguarded. In addition, all NWS have concluded AP's with the IAEA.

In the EU, the two NWS are under EURATOM safeguards. For these cases a distinction is made between peaceful and military activities. Also, gas centrifuge enrichment facilities are under safeguards in NWS. It is also expected that multinational facilities will be established for fuel cycle front end and backend services (multi-national approaches, MNA) and put under IAEA safeguards. In its annual safeguards implementation report (SIR) the IAEA informs about the evaluation results of its verification activities in NWS.

The last NPT Review Conference in May 2010 clearly stated the necessity for a fissile material production cut-off treaty (FMCT) with legally binding IAEA verification to ensure the irreversible stop of fissile material production. That would also lead to a general separation of peaceful and military activities in NWS, a further dismantling of military facilities and a wider application of IAEA safeguards in NWS.

The 2010 NPT Review Conference expected that, after achieving „Global Zero", comprehensive safeguards with AP's would become the universal safeguards standard in all states including NWS.

As already mentioned, the IAEA, with the further development of state level safeguards, will possess a powerful instrument to verify the correctness and completeness of states' declarations, i.e., to detect efficiently and effectively undeclared nuclear materials and facilities in states. This concept will also be of a big advantage in NWS.

For safeguarding complex military bulk handling facilities, the IAEA can rely on experiences from peaceful commercial facilities for the reprocessing of spent fuel, production of plutonium containing fuel, and uranium enrichment.

The verification of stocks of nuclear weapons or their dismantlement is more challenging, since it is necessary to protect sensitive or proliferation relevant information. Here, first experiences are available from the Trilateral Initiative (IAEA, Russian Federation and US), which has proposed the concept of an information barrier to protect sensitive information. Recently, the UK and Norway together with an NGO (VERTIC) have analysed and proposed verification elements for the FMCT.

5. Summary

The further development and implementation of state level safeguards has been asked by the 2010 NPT Review Conference. This new system combines the optimisation of inspection effort and efficiency since it offers a tool to focus effort in areas of concern. The decoupling of inspection effort from material quantities and qualities is a positive result of these developments.

The Agency has already shown considerable progress in implementing state level safeguards for the SIR evaluation. State specific reporting in the SIR appears more transparent and understandable.

The future evolution of state level safeguards offers also perspectives for implementing safeguards in NWS to replace voluntary offer safeguards or verification activities in connection with the FMCT.

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Implementation of Integrated Safeguards in Germany

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Abstract

Integrated Safeguards (IS) started in Germany on 1st of January 2010. It was preceded by a trilateral meeting between IAEA, EURATOM and the German Federal Ministry of Economics where the IAEA explained the new regime. Random interim inspections with a notification period of 24 h as a replacement of the classical quarterly interim inspections for spent fuel are among others the principal items in the new Safeguards regime. In the statement of the Federal Ministry of Economics generated in response to IAEA's proposals the operators' view was considered with the focus on legal basis of the new approach, sovereign tasks, feasibility of the IS-regime, joint team approach. The different views of IAEA and EURATOM with regard to the camera interval to be evaluated and the data to be transferred remotely increases the risk of divergence between the two institutions. The first experience with the new Integrated Safeguards regime will be presented. Also, the question whether the IS system will reduce the operators' effort for Safeguards activities will be addressed.

Keywords: Integrated Safeguards; first experiences; State-level approach

1. Introduction

The legal framework for the Safeguards-system in Germany is built upon chapter VII of the EURATOM Treaty and the Treaty on the Non-Proliferation of Nuclear Weapons put into practice in 1973 as the IAEA Safeguards agreement INFCIRC/153 which is the basis for all comprehensive safeguards agreements between IAEA and Non-nuclear weapon states (NNWS). In Germany as in all other NNWS members of the EURATOM Treaty the INFCIRC/153 entered into force in 1977 in terms of the verification agreement INCIRC/193. The Safeguards measures based on INFCIR/153-type agreements are commonly referred to as the "classical" safeguards system which verification measures were restricted to the facilities delineated in the design information. Under the classical Safeguards regime the state and the Community cooperate with the IAEA to provide evidence that the existing nuclear material and all nuclear activities are in line with the declaration.

With revealing the clandestine nuclear program of Iraq it was obvious that the classical safeguards system was not sufficiently designed to detect nuclear material and nuclear activities that had not been declared to the IAEA. In an effort to overcome the weakness of the classical Safeguards system the need for additional legal authority and additional Safeguards measures was recognized. The Safeguards strengthening measures include an extended access to information including reporting of nuclear imports and exports, extended access to locations in a state and the use of new and advanced verification techniques, in particular environmental sampling, which enables the IAEA to draw independent safeguards conclusions. Whereas a part of these strengthening measures could be implemented within the framework of INFCIRC/153 others required an extended legal basis as defined

in the additional protocol (AP) to the INCIRC/153 which was published as INFCIRC/540 in 1998. The Community signed the AP in 1998 which entered into force in the Community in 2004. It provides the necessary legal basis and tools for the IAEA to implement the key measures to confirm the absence of undeclared nuclear material and clandestine activities in the state as a whole.

A thorough state evaluation conducted by the IAEA and satisfactory answers to all nuclear fuel cycle related questions, including those concerning the past, are the basis for the credible assurance of the completeness of the declaration of the state. Thus the IAEA gets the knowledge of all nuclear fuel cycle related activities performed in the past and a comprehension of the current and planned activities. Consequently, the strengthened Safeguards measures enable an evaluation of the whole state to provide the basis for a state level approach instead of focusing on the facility level as performed under the classical Safeguards system. The broader conclusion is one of the prerequisites for such a state level approach which represents a holistic approach to the implementation of IS in a state. The broader conclusion for Germany was drawn by the IAEA in 2008 as stated in the Safeguards Implementation Report (SIR) for 2008. It needs to be renewed on an annual basis. Therewith, Germany reached the milestone indicating that the conditions for the implementation of integrated Safeguards (IS) are fulfilled. IS offers the possibility for an optimized combination of classical and strengthened Safeguards measures aiming at achieving the maximum effectiveness and efficiency without exceeding the existing resources but decreasing the required resources. Our expectation was that the application of IS would enable the IAEA to focus their verification efforts on the sensitive facilities assumed to represent a pronounced proliferation risk within the nuclear fuel cycle. For this reason we expected that the number of inspections will decrease in nuclear power plants and spent fuel dry storage facilities whereas the number of inspections in the sensitive facilities consisting in Germany of the uranium enrichment plant at Gronau and the fuel fabrication plant at Lingen will change to a lesser extent.

The paper describes the German preparations for the implementation of IS, the IS approach for Germany and deals with the divergence of the inspection regime put into practice by IAEA and EURATOM. The main focus is laid on the experience with the new IS regime during the first year after implementation taking into account the effect of the IS regime on the number of inspections and the inspection effort on the part of IAEA, EURATOM and operators.

2. German preparations for the implementation of IS

The adjustment of the present Safeguards system to the new Safeguards measures as required under the IS regime was not expected immediately after the submission of the initial declaration under the AP in the year 2004 but we did not expect it to take such a long time until the receipt of the Broader Conclusion. Between the beginning of 2005 and the end of 2009 the IAEA conducted around 40 complementary accesses (CA) in Germany and we received several formal requests for clarification according to paragraph 2.c.of the AP. The generic concept how to implement IS in the Community, i. E also in German LWR's, SFSF's, the FFP and the Uranium enrichment plant was outlined in IAEA/EURATOM-documents as a Partnership Approach under Integrated Safeguards. Consequently, these PA-documents represent an arrangement primarily made between EURATOM and IAEA with little involvement of the operators. These documents were signed in HLLC in June 2008. The following discussions led to the result that the new IS related inspection mode and activities were defined in the generic PA documents in a quite vague manner offering much room for interpretation due to the general wording. The operators had always requested that the new Safeguards measures be specified in the legal documents, the particular Safeguards provisions (PSP) and facility attachments (FA). These documents form the legal basis for carrying out inspections in a specific plant and are in our view a mandatory requirement for the implementation of IS. Even for many operating facilities PSP and FA have not been enacted yet. Furthermore the existing ones need to be updated. We think that updated PSP and FA have to be established as soon as possible.

On a trilateral meeting with representatives of the German authority, EURATOM and IAEA held in Berlin on 19th May 2009 IAEA gave a review how IS is to be implemented in Germany. In response, the German government announced a statement. Therefore internal German meetings with the participation of operators took place to discuss the IAEA's suggestions. The German interpretation of open issues was argued in two letters. The first one which had been sent to the IAEA and Euratom on 23 July 2009 comprised a general statement followed by a second letter in October 2009 with additional comments. A letter advising that IS was being implemented in Germany was received by EURATOM on 15th December 2009, thus setting the course for enacting the new IS regime from January 01, 2010.

3. The IS approach for Germany

The main changes in the IS approach compared to the classical approach stem from the extension of the timeliness goal for irradiated fuel from three months to one year and an improved emphasis on unpredictability. Therefore the key item of the IS regime is the transition from announced quarterly interim inspections to randomly performed interim inspections (RII) with a probability of 20% per year and a preannouncement period of 24 h.

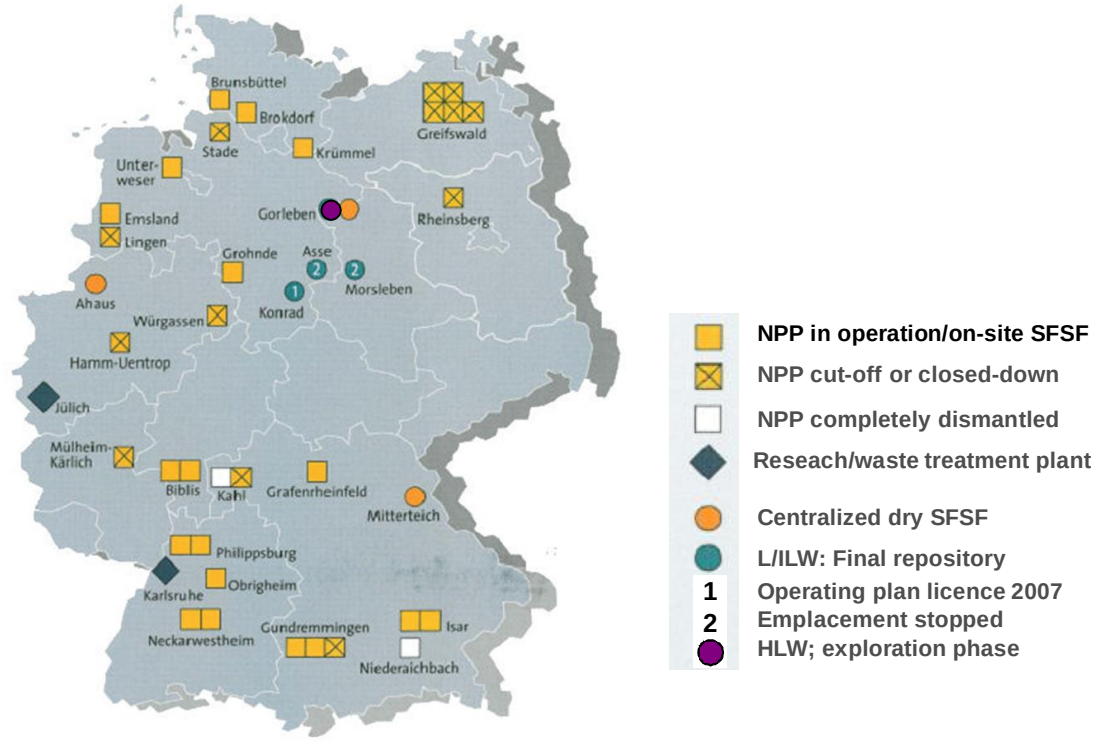

Figure 1: Nuclear power plants and spent fuel storage facilities in Germany

3.1 LWR

In the above mentioned meeting with German government officials and Euratom in May 2009, the IAEA presented their concepts for introducing Integrated Safeguards in Germany. At that time, a total of 27 nuclear power reactors were under IAEA safeguards in Germany, 17 of them were operational and 10 of them permanently shut down or closed down. Since IS affects the operating plants only four SNRI could be assumed for reactors.

Apart from the discontinuation of the quarterly interim inspections and their replacement by a random interim inspection (RII) with an advance notification of 24 hours to the operator the advantages that the operators saw in the presented IS concept were on the one hand a facilitation in the conduct of a PIV as a particular core control is no longer required and on the other hand a different role of surveillance. The attention of the video surveillance is paid to the 24 h interval between inspection announcement and arrival of inspectors thus enabling a continuous video surveillance in overwrite mode. In contrast to traditional Safeguards the focus during PIV was shifted to pre- and post-loading inspections by cancelling the open core inspections and limiting Safeguards to video surveillance during the open core period provided that no SF transfers occur during the reloading period. The core is sealed at the post-loading inspection. The IAEA stated that surveillance failures outside of these time periods would not require any re-verification activities from their side. From the past it was known that surveillance may suffer from many failures or malfunctions that lead to requirements for re-verification to re-establish the continuity of knowledge. Therefore this IAEA statement was well recognized.

Fresh fuel is verified for gross defects at pre-PIV and item-counted at post-PIV whereas spent fuel is verified for gross defects twice at pre- and post-PIV as well. For reactors with fresh MOX fuel on inventory, the IS concept foresees a reduction in the number of annual verifications, the timeliness goal is reduced from one month to three months and the number of inspections from 12 to four due to extended timeliness goal for MOX fuel from one to three month With regard to the receipt of fresh MOX assemblies, IS does not provide for changes in the inspection requirements.

The main inspection effort concerns the transfer of spent fuel from the reactor to the interim storage facility. The inventory control of the loaded Castor casks prior to cask closure is omitted under the IS regime so that currently the cask sealing after loading is the only inspection performed by IAEA and EURATOM between pre and post campaign verification. Aiming at decreasing their inspection effort further the IAEA is interested in a cask sealing performed only by the operator or EURATOM since thereby IAEA expects to save 75 % of their inspection effort which is caused by the about 40 annual CASTOR cask loadings due to 17 operating reactors. The IAEA would like the operator to apply an electronic seal under video surveillance. With the lack of any verifiable confirmation that the seal has been applied correctly there would be no way for the operator to recognize whether he accomplished the sealing procedure correctly or not.

3.2 Spent fuel storage facilities

Since mid-2005, any transport of spent fuel to reprocessing abroad has been abandoned, the current policy restricts the disposal of spent fuel elements exclusively to direct final disposal. Until a final repository becomes available intermediate storage is the only way for nuclear waste management in Germany which occurs in central dry storage facilities and decentralized on-site interim storage facilities close to the reactor. .

The IAEA assigns the existing storage facilities into two groups, group one are facilities with an inventory of one or more significant quantities (SQ) of direct use material which counts 16 facilities and the second group consists of 3 facilities with an inventory between 0.5 and 1 SQ. The IS concept for the 16 facilities in group one foresees an annual PIV and a random interim inspection (RII) with an advance notification of 24 hours to the operator and a 20% selection probability per year per storage installation. In group two, one PIV is foreseen in 4 to 6 years on the average.

In consideration of 16 SFSF with more than one SQ including 12 on-site dry storage facilities, the two central storage facilities Ahaus and Gorleben, the ZLN, the dry storage facility at Jülich as well as the wet storage at Obrigheim no more than 4 SNRI could be expected per year. That means one RII can be assumed in the same facility every five years on average. Including the reactors that means that in Germany the annual number of RII is 8 in total. The annual PIV per storage remained unaffected.

Here again, the reduction in the number of inspections as derived from the concept can only be realized in completely static situations. As soon as there occur movements or inventory changes or maintenance work on storage casks the picture may change considerably.

3.3 Fuel fabrication and enrichment facilities

With regard to fuel fabrication plants (FFP) and gas centrifuge enrichment plants (GCEP), the IAEA introduced, except for the transition from announced and planned inspections to RII, a further new item in the safeguards concept, a mailbox system. According to an agreed schedule, the operator deposits specified near term information relevant for verification activities in a computer system accessible by the IAEA. This can be information on planned production, shipments and receipts as well as updates of such information in the case that the realized activities deviated from the planned activities or the schedule had changed. This information helps the IAEA in inspection planning and determining their verification activities during RII.

Irrespective of the introduction of IS, the safeguards approaches for FFPs and GCEPs were revised by the IAEA to cover a wider range of diversion respectively misuse scenarios so that a direct comparison of the situation before and after the implementation of IS is not possible. In contrast to reactors and storage facilities FFPs and GCEPs require individual adjustments of their safeguards concepts to plant specific characteristics. Therefore for FFPs and GCEPs the procedure of implementing IS was different from that applied in reactors and storage facilities. Correspondingly, For the German fuel fabrication plant in Lingen as well as for the enrichment facility in Gronau special agreements between the IAEA, Euratom and the operator were made on how to implement the revised safeguards approaches under IS.

4. Operators' views and experiences

On the trilateral meeting BMWi agreed to comment the IAEA concept for IS implementation in Germany. The IAEA concept was also examined by operators of the nuclear fuel cycle who joined the meeting at BMWi on 10th June 2010. They defined their position regarding the following topics:

1. legal basis for IS
2. Joint team approach
3. Practicability of the new Safeguards measures
4. Handling of CASTOR-cask sealing
5. Effort for the operator

4.1 Operators' views

Since the FA remains the legal basis in the new IS system the operators emphasize the adjustment of the FA's and PSP or the generation of the corresponding documents to reach legal certainty as soon as possible. For the operators is the continuation of the joint team approach regarding inspection content and evaluation as well as the practicability of the new Safeguards measures of essential importance. The operators regard the sealing of CASTOR casks as a sovereign task which the operators are not willing to perform especially since the cask sealing is regarded under IS as a more critical step due to the abolition of inventory control prior to cask closure. Cask sealing by the operator might only be taken into account if the sealing is subordinated as a back-up solution. Besides that the potential risk of an improper application associated with a negative impact on the public awareness is not acceptable for the operator in Germany.

For the acceptance of the new IS-regime it is important that the new Safeguards measures do not only lead to a reduced effort for the IAEA but also to a noticeable decrease of the effort for the operator. In this context the operators underline the importance of two aspects:

- The claim of the IAEA to trigger follow-up inspections in case that the inspection goal has not been reached or necessary equipment is not available so that the RII cannot be performed as planned.
- To what a degree IAEA and EURATOM pursue one inspection regime in principle, since under the new IS regime IAEA and EURATOM set a different focus. The operators fear an increase of their efforts caused by independently performed Safeguards activities due to a different task definition between IAEA and EURATOM. Accordingly, the IAEA focuses on the detection of non-declared nuclear material and activities by evaluating only the preannouncement period of 24 h whereas EURATOM is interested in the complete video recording round the clock to preserve the continuity of knowledge. A divergence of the inspection regime is also reflected by the topic “remote data transmission”. For the detection of non-declared nuclear material and activities the transmission of health data which indicate the functional status of a video camera during the preannouncement period of 24 h should be sufficient whereas EURATOM also need the transmission of seal states and Safeguards image data to use RDT as a planning tool for inspections. In view of the appearance of the Stuxnet virus in 2009 data security remains a main challenge which has to be met.

4.2 Experience with the new IS regime during the first year after implementation

Eight RII in reactors and storage installations have been performed in 2010 in total in Germany so that the number of RII was in line with the statement made during the trilateral meeting. Based on the wording of “at least four” as mentioned in the PA-paper for reactor or SFSF a higher number of RII than the average could not be excluded initially. All RII were accompanied by EURATOM. The reactor Obrigheim which was shut-down in 2006 was attributed to the operating reactors since the reactor is still not free from spent fuel assemblies. No RII gave cause for complaints so that all RII were completed successfully. Also, the redundant pre-announcement of RII to the operator by EURATOM and the federal state authorities by means of a fax on the day before the inspection day occurred without problems.

Date of RII	Facility	Location	2010
23 Feb	Dry storage	Emsland	First half of the year
09 Mar	Reactor Block A	Biblis	
23 Mar	Dry storage	Biblis	
20 Apr	Reactor Block 1	Philippsburg	
28 Apr	Reactor Block A	Biblis	
04 May	Reactor	Obrigheim	
25 Aug	Dry storage	Isar	Second half of the year
29 Nov	Dry storage	Gundremmingen	

Table 1: Random interim inspections performed in Germany in 2010

Yet, it is noticeable that the RII accumulated in the first half of the year 2010 within the two months March and April. Accordingly, the last of the four in the reactor orderly performed RII was already accomplished within the first half of the year on 04th May. Also half of the dry storage related RII

occurred within the first half of the year so that only two of the eight RII were performed in the second half of the year. After a summer break of nearly four months the seventh RII occurred in August and the last one in November.

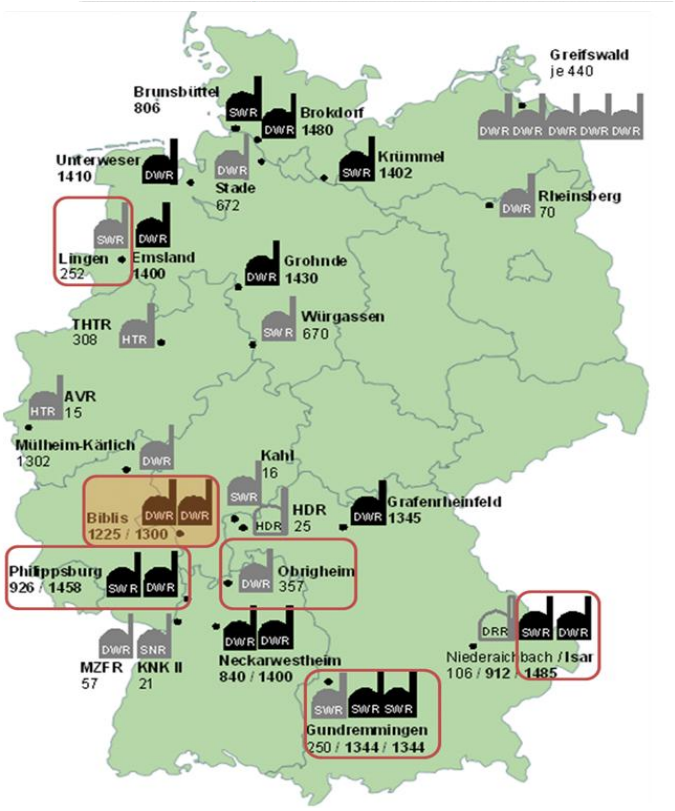

Figure 2: Occurrence of random interim inspections

Furthermore the accumulation of RII with the exception of that in Emsland in the southern half of Germany is striking. In addition to that three out of the eight RII occurred in the same location at Biblis. Within one and a half month two RII were performed in the reactor block A and the third one in the on-site dry storage facility. It is astonishing that such a spatial and temporal accumulation of RII is the result of a statistically based selection procedure as required for RII. Finally it should be mentioned that no RII was combined with a complementary access.

Taking a closer look to the inspection effort it is obvious that the IAEA succeeded in reducing their effort substantially due to a reduced inspection frequency caused by the replacement of routine inspections by random RII. However, a reduced IAEA inspection effort does not necessarily result in an equally large decrease of effort for EURATOM or a noticeable decrease for the operator at all. This may be due to different reasons:

- Extra EURATOM-inspections occurred in the absence of IAEA in Biblis, Philippsburg and Gorleben and were performed especially in the first quarter of 2010. EURATOM explained these additional inspections with the closing of the traditional regime at the end of 2009. Besides that, surveillance equipment needed a greater memory capacity e. g. to save a one-year continuously ongoing camera recording between two PIV per material balance zone (MBA).
- The operators are now faced with the permanent need to up-date the documents for accountancy near real-time and to balance the lacking staff in case of vacation or illness in order to be always prepared for the RII. Besides that the operator now has to announce all incidents timely which may have an effect on RII performance in order to avoid follow-up inspections.

1. It is the type of installation which determines the extent of efforts for the operator. In the two central storage facilities that have a static inventory the effort is limited to the assistance of the operator during the PIV and RII. Therefore the decreasing number of inspections from a total of eight consisting of two PIV and six routine inspections to three inspections including one EURATOM-inspection became also obvious for the operator. In contrast, there was no noticeable decrease of effort in case of reactors and on site storage facilities although the inspection frequency decreased from approximate 200 to 145 for the reactors and from app. 50 to 30 for the on-site storage facilities acc. EURATOM. This impression may be due to the observation that a clear separation of usual random inspections under IS from scheduled inspections linked to CASTOR-cask sealing was not possible especially since in view of about 40 CASTOR-loadings per year those inspections make up the biggest portion of the total inspections.

5. Conclusion

In conclusion, we can summarize our experiences as follows

- The first year of application of Integrated Safeguards did not show any major difficulties in the conduct of the inspections. But of course, after just one year this can be a preliminary conclusion only.
- For a solid implementation of IS in German facilities, a solid legal basis is needed. The operators insist in having up to date facility specific legal documents for safeguards implementation which means up to date PSP and FA for all facilities.
- The operators strongly support all activities to strengthen the global non-proliferation efforts and contribute to that effort at their best. The expectation with IS however were that besides effectiveness also the efficiency of safeguards would be increased and that the state level approach would reduce the burden for the single operators. At present, this can only be seen to a small extent and only under specific conditions.
- IS is a concept applied to IAEA safeguards. If IAEA safeguards approaches and Euratom safeguards approaches are not harmonized to the greatest possible extend, the burden on the operator will be unnecessarily high. IAEA and Euratom should undertake every sensible effort to make their inspection procedures mutually compatible to reduce the overall burden on the operator.

Safeguarding a Waste Treatment and Storage Installation – An Operators View

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Abstract:

In former times safeguards rules for waste treatment and intermediate storage facilities were not officially defined. The existing policy papers were only partly known to the operators. Due to this fact, the structure of such a facility did primarily consider the needs for safe operation but not for safeguards. In EURATOM Regulation 302/2005 a specific questionnaire for these facilities has been incorporated. This paper describes the challenges, for the operator as well as the safeguards inspectorates, in safeguarding the central decontamination department (Hauptabteilung Dekontaminationsbetriebe - HDB) of the Wiederaufarbeitungsanlage Karlsruhe Rückbau und Entsorgungs- GmbH (WAK).

HDB has been built in the late 1950 as part of the former Nuclear Research Centre Karlsruhe in order to dispose of radioactive wastes arising from the Karlsruhe research facilities. For this purpose HDB operates facilities for treatment of liquid and solid raw wastes which may be combustible or not. The task is to decontaminate radioactive components as well as to condition and store low and intermediate raw wastes and waste products. Since the early 1990 HDB has reported to EURATOM quarterly using an informal system and the facility was inspected by EURATOM on a regular basis. With Regulation 302/2005 coming into force, Basic Technical Characteristics (BTC) are documented and monthly Inventory Change Reports (ICR) are given. In 2009 IAEA inspected HDB for the first time.

HDB soon has recognized several limitations in fulfilling safeguards requests. Regarding intermediate raw waste and waste products, for operational reasons these items are stockpiled close together and therefore hardly to measure. Due to high fission product activity and low concentration of fissile material, a verification of the declaration is in general almost impossible. To meet the requirements of customers and authorities enriched nuclear material may be blended prior to processing. Furthermore, raw wastes of different origin are mixed and treated together in some cases. Therefore, charge tracing makes no sense.

Considering these limitations, safeguards basically has to rely on verifying the BTC and the internal declaration rules of the operator. As the results of the nuclear material accounting at HDB can be compared with those of the deliverers, the values have to be acceptable in this context.

Keywords: waste treatment; safeguards limitations;

1. Assignment of HDB

The Central Decontamination Department (HDB) of the WAK GmbH is the central facility for the treatment and conditioning of radioactive material at the Karlsruhe site. HDB is specialized in handling radioactive wastes and residues from decontamination and recycling to conditioning for final storage. The department provides waste disposal services for all nuclear research reactors and the former nuclear fuel reprocessing plant at the site, as well as for the European Institute for Transuranium Elements, for State Collection Centres and for external clients.

Radioactive wastes are treated and conditioned for harmless recovery or a later transfer to a final repository. For this purpose, following methods of treatment and conditioning are used:

- decontamination, compaction, cementation and incineration of solid wastes of low and intermediate activity,
- evaporation and incineration of liquid wastes.

Central Decontamination Department

By disassembling and decontamination, one part of the delivered components can be released as reusable materials. The other parts are treated as radioactive wastes and prepared to meet the regulations for final storage. The conditioned radioactive casks remain in HDB interim storage facilities until delivery to a final repository, or are returned to the owner.

2. Conditioning Facilities

2.1. Decontamination

With decontamination, radioactive material is removed from the surface of dismantled plant components. The procedure's objective is to decontaminate plant components to the point where they can be released from radiation protection control or melted, and will not have to be conditioned for final storage. Depending on the nature of the plant components and the contamination, there are several possible decontamination methods:

- physical procedures such as steam or sand blasting,
- mechanical surface removal such as grinding, planning, milling and turning,
- chemical procedures such as chemical stripping or leaching,
- physical-chemical procedures such as dry ice blasting.

Most of these tasks are performed in caissons with separate ventilation systems, which are only entered by personnel wearing gas-tight protective suits.

2.2. Low Level Waste Scrapping and Compaction

All material and components which cannot be decontaminated have to be disposed of as radioactive wastes. Therefore, their volume is reduced as far as possible. All solid, unburnable wastes with a surface dose rate up to a maximum of 2 mSv/h are transferred to the LLW Scrapping Division.

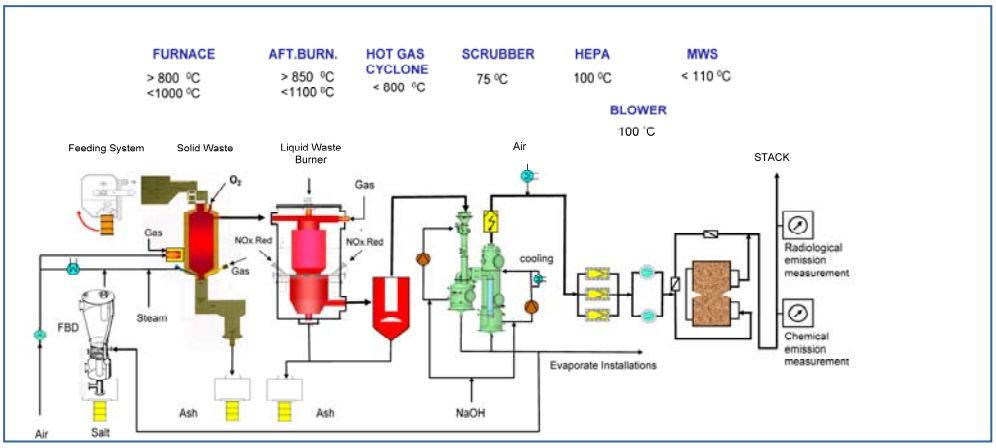
In the scrapping caisson, thermal cutting or manual disassembling is performed. The preliminary press has a compaction force of 5000 kN. The preforms are packaged into 170-l-barrels and then proceeded

to further reduction in a high-pressure compaction system with a compaction force of 15000 kN. The LLW scrapping achieves a volume reduction of a factor of 6 in total.

2.3. Incineration

The incineration facility is constructed for solid and liquid burnable wastes with low radioactive contamination. Solid wastes are collected in foil bags or cardboard boxes, brought into the control box via a double-lid system, and transferred to the furnace. Liquid wastes are delivered to HDB in collection tanks, analyzed for activity and halogenated hydrocarbons, and, in batches, transferred to incineration in the afterburning chamber.

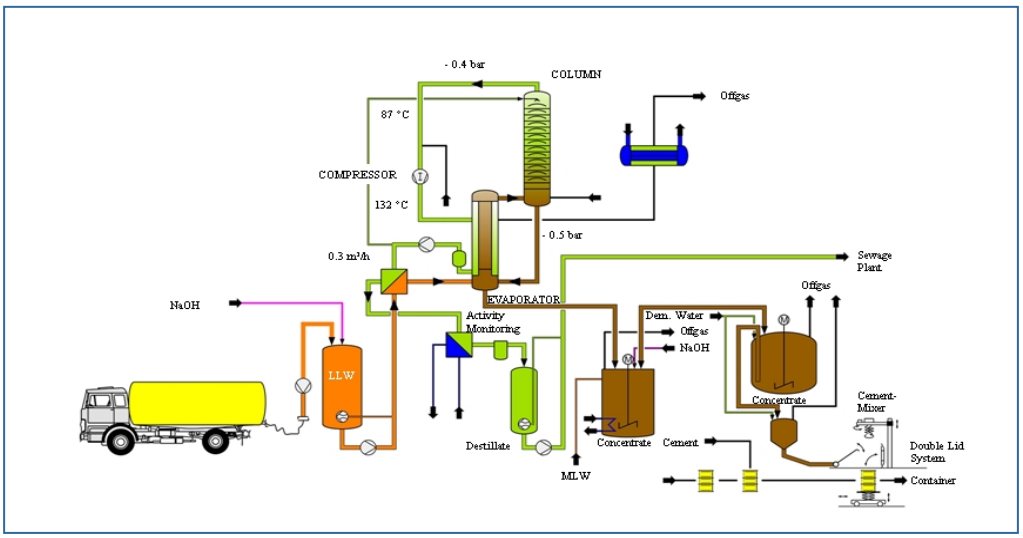
The radioactive pollutants in the wastes end up either in the ash or in the incinerator's exhaust gas. The ash is filled into barrels and then high-pressure-compacted. The exhaust gas is fed into a multi-level cleaning system with scrubbers, absorbers and filters. Due to this method, both activity discharge with exhaust air and emission of chemical pollutants can be reduced to values far below the limits specified by the authorities. Treatment in the incineration facility followed by compaction of the ash reduces the wastes' volume by a factor of up to 100.



Flowchart of the incineration facility

2.4. Evaporation and Solidification

Liquid inorganic wastes, such as chemical effluents, are evaporated. The pollutants are concentrated in the evaporation residue, with only a very small proportion ending up in the distillate. In order to condition the wastes for final storage, the evaporator concentrates are solidified into drums by homogenous mixing with cement.



Flowchart of the evaporation and cementation of the concentrate

For treatment of low active liquids, HDB operates a facility for evaporation and one for cementation. The distillate created by the evaporation is sampled, analyzed, and, after clearance, disposed of as sewage.

2.5. ILW Scrapping

“Intermediate-level waste“ (ILW) components are disassembled, filled into drums and then forwarded to high pressure compaction. Due to the wastes’ activity inventory and dose rate, crushing and compacting is performed in a remote-controlled manner. For treatment of the components, the facility has several “hot cells” varying in sizes and equipment. The personnel are placed outside these cells and operate the manipulators looking through stained glass windows, or via camera. Mechanical, low-emission procedures such as cutting, pressing or sawing are used.

Two cells are built right next to a loading/unloading cell. The cells have a direct transport container lock for wastes in special drums, as well as a double-lid system for 200 l and 400 l-drums.

Large working cell

Remote-controlled operation of a cell

The working cells are equipped with a drum gripping unit, a heavy-duty manipulator, and two master-slave manipulators at each cell window. For conditioning of wastes, hacksaws, a hydraulic cutter of 800 kN cutting force, and a hydraulic compaction unit of 20 000 kN compaction force are available.

3. Packaging and Storage Facility

Packages conditioned for final storage are held in interim storage at HDB, pending delivery to a repository. The storage capacity amounts to 77.000 m³.

German waste management concept requires two different kinds of repositories, namely one for heat-generating waste and one for waste with negligible heat generation. The latter usually store low and intermediate-level waste, which is quantitatively the majority of all conditioned casks. Germany is developing the repository KONRAD near Salzgitter for these wastes.

Storage requires casks in cylindrical drums and square containers with a maximum weight of 20 Mg each. Depending on the wastes’ radiological condition, packaging requires different materials with various insulation thicknesses. When choosing the appropriate packaging, it is imperative that the maximum permitted dose rate does not exceed 2 mSv/h on the casks’ outside surface. Drums have to be licensed for reposition. At HDB, casks are prepared for repositioning in KONRAD. This includes measures for product control as well as the sealing of the drums.

4. Analytical Service and Documentation

4.1. Analytical Service

Meeting guidelines stated by the authorities and repositories requires extensive measurement techniques. HDB applies:

- a multi-purpose radiochemical laboratory for analysis of radioactive samples ranging from low active to intermediate active,

- a variety of drum measurement systems for non-destructive control of residues and conditioned waste products delivered to HDB and
- a release measurement facility for release of decontaminated waste according to § 29 Radiation Protection Regulation. [1]

At the radiochemical laboratory, various methods of nuclear radiation measurement, radio- and chemical analysis are applied.

For delivery- and product-control, a neutron monitor for the evaluation of the fissile material inventory and in-situ measurement systems for γ-spectrometric examination of various geometrical shapes are available, next to a variety of drum measurement systems. They allow for an investigation of dose rates from a few µSv up to the Sievert range.

LLW-drum measurement

Neutron monitor

Release measurement facility

A release procedure according to § 29 of the Radiation Protection Regulation [1] is based on the results of the release measurement facility and is operated by the safety department. These measurements require that nuclide vectors of wastes are known, because the activity of indeterminable nuclides has to be considered arithmetically. The facility is based on the principle of total gamma measurement, gamma quanta of more than 200 keV are measured. It is mainly used to measure bulk material, due to the impossibility of direct surface measurement. The material to be measured is brought into the facility in 200-l-drums, or in grid boxes with a maximum capacity of 450 kg or 900 kg.

4.2. Documentation

§ 78 of Radiation Protection Regulation [1] requires the obligation for accountancy when handling radioactive wastes. One of its demands is the retraceability of the material from repository to the original waste producer. Therefore, all material flows have to be recorded and simultaneously assigned to the corresponding batch.

Using a data base, the specifically designed software **KADABRA** (**Ka**rlsruher **Da**ten**b**ank für radioaktive **R**eststoffe und **A**bfälle – Karlsruhe data base for radioactive wastes) collects and documents all relevant data according to the valid acceptance criteria, such as:

- Type of waste
- Nuclide inventory
- Owner
- Physical state
- Chemical composition
- Type and inventory of fissile material.

All conditioning steps are recorded and documented. The compliance with all legal regulations is monitored. All necessary reports are generated on the basis of this data base.

5. Flow of Material

5.1. Delivery of Radioactive Waste

- Preliminary check of documentation

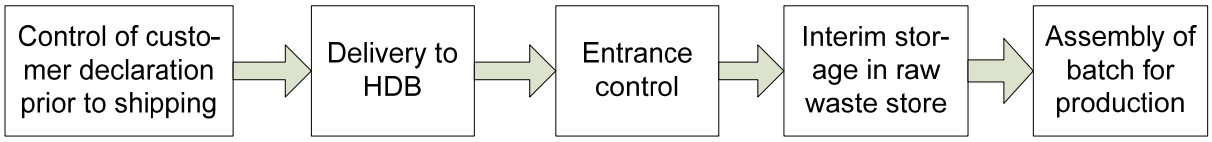
The Radiation Protection Regulation requires the obligation for accountancy when handling radioactive wastes. Furthermore waste acceptance criteria based on facility specific requirements and activity limits have to be observed, when delivering radioactive waste to HDB. For this reason the waste has to be specified on an individual dispatch note, which has to be subjected to a preliminary check at HDB prior to waste delivery.

- Entrance control

Approximately 10 % of the incoming material is subjected to an entrance control by non destructive assay (weight and dose rate measurement, γ-spectrometry and passive neutron monitoring). If discrepancies are detected the material is locked for processing and the customer will be consulted. If the customer's declaration lies within the defined tolerance limits, his data are booked and the material is approved for processing.

- Storage for Conditioning

After passing the entrance control the waste is transferred to the LLW or the ILW storage depending to the dose rate and activity inventory.



5.2. Conditioning

The waste is generally treated in batches, which are planned and assembled on the base of similar materials and nuclide inventories. If possible, the wastes of individual customer will be treated in separate batches. The batches are planned using the database KADABRA with programs checking:

- The allowed nuclide inventory of the facility.
- The release of volatile nuclides during production such as H-3, C-14 and I-129.
- The KONRAD acceptance criteria. [2]

The conditioning of waste products has to be defined by plans of procedures, which have to be approved prior to production by the Technical Inspection Authority on behalf of the Bundesamt für Strahlenschutz. In this process control quality plan the procedural steps and the parameters are defined according to the KONRAD acceptance criteria. The procedure of activity calculation has to be defined in this plan. The procedure depends on the type of waste material, the treatment during conditioning, the nuclide inventory and special requirements defined by the customer or the authority in charge.

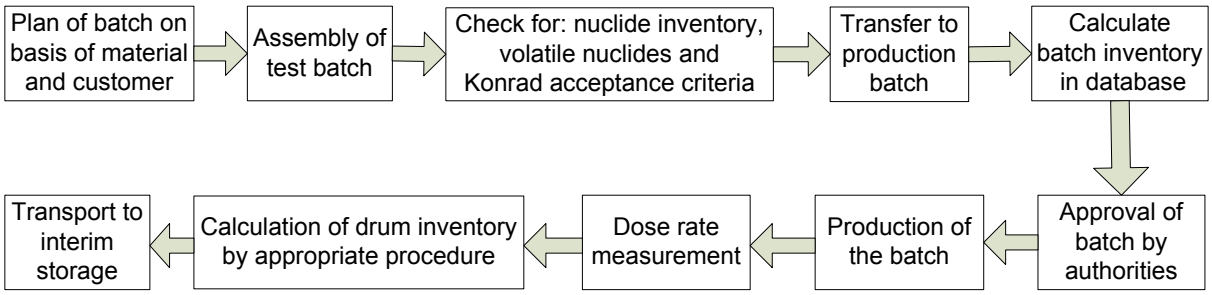
6. Waste Treatment Processes

6.1. Compaction in the Scrapping Facility

Waste dedicated for the scrapping facility is generally delivered to HDB in compaction drums. The batch is assembled on basis of similar materials and if possible sorted by customer. The activities of the declared raw wastes are booked via KADABRA into a test batch to plan the conditioning in accordance with the KONRAD acceptance criteria. If the batch is approved, the test batch is transferred into a production batch, which has to be approved by the production manager and the authorities.

The batch is processed in the scrapping facility (see 2.2). After production the pellets are transferred into 200 l drums. The amount of pellets, their weight and their nuclide inventory are assigned via

KADABRA to the products. The nuclide inventory is calculated on basis of dose rate measurements and the nuclide inventory of the batch (see 7). After dose rate measurement and declaration the drums are transported to the interim storage facility. There the product control is applied including second dose-rate measurement, photo documentation of the drums and γ-spectrometry to verify the declaration.



6.2. Evaporation and Solidification

For the acceptance of liquid waste a sample has to be analyzed by HDB to check the declaration and the chemical composition (especially tensides and decontamination fluids as well as acids have to be analyzed precisely to plan the batch in the evaporator). The resulting concentrate of the batch and the amount of distillate are calculated by the laboratory on base of the raw material analysis and checked for the production criteria.

After approval the batch is evaporated and yields distillate, which is after analysis transferred to the conventional sewage plant, and the concentrate, which contains the radioactivity. The concentrate is analyzed prior to solidification. The analytical results are used for the declaration together with the vector produced by summing up the declared nuclide inventories of the raw waste waters.

The concentrate is solidified in a batch by stirring into cement. The solidification properties are analyzed prior to production and monitored during production. After complete solidification the dose rate and the homogeneity of the nuclide distribution in the drum are analyzed in a drum measurement facility. The nuclide inventory of each drum in the batch is calculated in KADABRA using the analysis, the nuclide composition of the batch and the amount of concentrate per drum (see 7).

6.3. Incineration

Solid combustible wastes are booked into an incineration batch, which monitors the production criteria. After incineration the ash is sampled and analyzed for the nuclide inventory. The actinides are determined after chemical separation by mass spectrometry. The calculation of the nuclide inventory of the ashes for the batch is calculated in the database on the basis of the analysis and the vector produced by the summation of all raw wastes in the batch. The ash is supercompacted in the scrapping facility. There the nuclide inventory of the batch is divided to the individual waste products relative to dose-rate or mass, either, or recalculated on the basis of dose rate measurements. Another waste stream of the incineration facility, the scrubbing water, is dried to result in salt as a waste product for final storage.

7. Calculation of the Nuclide Inventory of the Waste Products

KADABRA allows several approved calculation procedures of batch nuclide inventories and individual waste products. The most common procedures are:

- Calculation of nuclide inventory per drum on the basis of *dose-rate measurements*:
  a) The nuclide inventory of the batch is distributed to the individual drums in relation to their dose-rate values.
  b) The nuclide inventories of the waste products (drums) are recalculated on the basis of their dose-rate values and the nuclide composition (nuclide vector) of the batch.

  These procedures are normally applied for large batches of compacted waste.

- *Pellet tracking*: The declared inventory of each pellet in a batch is recorded and the inventory of a waste product is calculated by the sum of the pellets in the drum.
  This procedure is especially designed for the compaction and is applied if the content of each pellet has to be declared.
- Calculation on basis of *analysis*, *batch inventory* and *mass of the waste product*: Detectable nuclides are analysed in the laboratory by different analytical methods as α-, γ-spectrometry, etc. Not detectable nuclides are added from the batch vector related to a defined key nuclide (e.g. Co-60, Cs-137). The obtained mass specific results are used to declare the nuclide activities of the waste products on basis of their individual mass (e.g. concentrate added in each drum for solidification).
- Calculation on basis of *analysis*, *batch inventory* and *dose rate*: The nuclide composition is determined by analysis and correlation as described before. The inventory is assigned to the individual drums via dose-rate values.
  This method is applied for the calculation of the inventory of bottom ash from the incinerator, which is supercompacted in a second step.

In addition to the nuclide inventory resulting from the processed waste in the individual batch cross contamination from batch to batch has to be considered. This cross contamination is calculated as a defined small fraction of the batch inventory and is transferred to the subsequent batch.

8. Fissile Material Accounting

Nuclear material arriving at HDB in the form of waste (material code “W”) is booked into the Material Balance Area (MBA) with code “RD”, entered into the database and reported monthly by the Inventory Change Report (ICR), which is extracted from the database. When the material is transferred to conditioning, it is removed from the MBA by booking as “TC”. The incoming mass of fissile material corresponds to the mass, which is transferred to production. During the conditioning the activities of the radio nuclides and consequently the masses of fissile materials are recalculated on the basis of analytical results or dose rate measurements. Due to blending of enriched nuclear material prior to processing or the combined processing of different materials the kind and grade of enrichment may change. For this reason the waste products often do not correspond to the raw wastes.

The generation of waste products containing fissile material is reported to EURATOM once a year according to annex XIV of EURATOM Regulation 302/2005. [3]

9. Boundary Conditions for Safeguarding Waste

At HDB nuclear material is accepted in the form of radioactive waste exclusively. The nuclear material is available in a form and dilution that is not suitable for reuse. It is mixed with fission and activation products and usually contained in a waste matrix. Nuclear material that is delivered from an external MBA to HDB is randomly checked for its nuclide inventory by non destructive analysis. Afterwards it is transferred to the entrance storage facilities (LLW or ILW storage). These storage areas represent the entrance MBA which is totally under EURATOM and IAEA safeguards.

In this MBA the incoming nuclear material is stored along with other radioactive raw waste and intermediate products in piles of drums or container waiting for processing. In these piles the material of interest is not accessible for control measures with justifiable effort. In the ILW storage facility the waste is stored in hot cells together with hundreds of conditioned waste products of dose rates up to some 100 Sv/h. This storage area cannot be entered for control purposes. To facilitate the (at least partly) identification of items during Physical Inventory Verification (PIV), HDB is able to provide additional listings to the inspectorates.

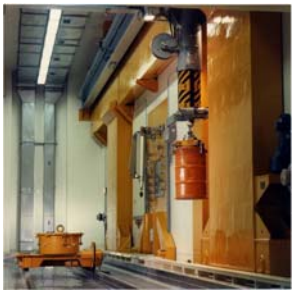
Concerning the non destructive analyses, γ-spectrometry indicates the activity of fission and activation products, mainly Cs-137 and Co-60. Uranium and plutonium isotopes can hardly be detected in radioactive waste, because of the low gamma energies and the high detection limits (due to a low transition probability) of these isotopes related to the fission and activation products. As far as the passive neutron monitor is concerned, amounts of > 10 - 100 mg of plutonium can be detected in radioactive waste, depending on the isotopic composition, the chemical speciation and the waste matrix. In con-

trast, the detection limit of pure uranium is in the order of some kilogram. Pre-requisite for an accurate appraisal of the analytical results is the knowledge of the definite nuclide and isotopic composition as far as neutron emitter are concerned. Especially the presence of curium and californium, often not declared, may lead to an overestimation of fissile material, for instance plutonium.

During the conditioning processes the nuclear material may be mixed to achieve a product meeting the requirements of the final repository. According to the specific process control quality plan the once declared U- and Pu-masses are propagated. Although measurements often do not lead to satisfactory results, the nuclear material balance of HDB is transparent. Furthermore, the nuclear material input of HDB must be an acceptable waste output of the sending nuclear facility.

Last but not least, EURATOM and IAEA have the possibility and the right to verify the BTC of HDB. That means the inspectorates are able to verify the absence of a diversion path, because an installation for recovering fissile material is missing.

LLW storage



ILW storage facility

10. Conclusion

Founded more than 50 years ago the central decontamination department (HDB) is a nuclear installation which was not designed for fulfilling safeguards obligations. Due to the fact that the German Radiation Protection Regulation and the requirements of the coming final repository KONRAD have increased within the last 20 years, HDB today has an improved knowledge about the nuclear material content in waste. Providing additional listings to the inspectorates during PIV, it is possible for them to identify (at least partly) items from the PIL. Although, independent measurements often are impossible, the design of the plant and the operational and accounting procedures are transparent. By verifying these circumstances the inspectorates are able to draw the conclusion of non diversion.

11. References

[1] *Verordnung über den Schutz vor Schäden durch ionisierende Strahlen (Strahlenschutzverordnung) vom 20. Juli 2001, BGBl. I, p.1714*

[2] *Anforderungen an endzulagernde radioaktive Abfälle (Endlagerbedingungen, Stand: Dezember 1995) - Schachtanlage Konrad - , Bundesamt für Strahlenschutz, ET-IB-79, Salzgitter*

[3] *COMMISSION REGULATION (Euratom) No 302/2005 of 8 February 2005 on the application of Euratom safeguards, Official Journal of the European Union, L54, 28.2.2005, 70 p.*

09 Destructive Analysis measurements

The Future Role of the IAEA's Network of Analytical Laboratories

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Abstract:

The IAEA's Network of Analytical Laboratories (NWAL) is a mature initiative that has grown to meet the IAEA's requirements and those of the safeguards analytical community. It currently consists of nineteen laboratories in nine Member States, the European Commission and the IAEA, providing reference materials, quality assurance programmes, bulk and particle analysis of environmental samples and the analysis of nuclear material samples. The capacity and capability of the network is expanding with new laboratories, new technologies and new facilities. The manner in which the network is developed and maintained should be reviewed to reflect this expansion. This paper outlines a history of the NWAL to-date, shows the current status of the network in each of its component areas, highlights the future requirements/issues in each area, and suggests how to meet the challenges. A number of long-term programmes are suggested for Member States and the IAEA to pursue, providing partners with new responsibilities that will enable further progress in the analytical work.

Keywords: environmental; network; destructive; analysis; safeguards

1. Introduction

As part of the Nuclear Verification Programme at the International Atomic Energy Agency (IAEA), samples collected by IAEA safeguards inspectors are analyzed to help determine the completeness and correctness of a Member State's declarations. These analyses are performed by the IAEA's Safeguards Analytical Laboratories (SAL), located in Seibersdorf, Austria, and by a Network of Analytical Laboratories (NWAL). Members of the NWAL are nominated by their governments and support the IAEA by providing expertise, performing sample analysis, and supplying reference materials.

The NWAL has four different components: environmental sample analysis, reference and quality control materials, nuclear material analysis, and heavy water sample analysis.

2. History of NWAL

The idea that the IAEA should possess its own safeguards analytical services was conceived in 1970, when the Non-Proliferation Treaty (NPT) entered into force, and the IAEA Safeguards Committee was established [1]. At this time (and even before), IAEA inspectors were collecting samples of safeguarded nuclear material for chemical and isotopic analysis. Laboratories in several Member States performed the analyses through *ad hoc* arrangements between the Member State and the IAEA. [2]

By 1976, a fully staffed and equipped Safeguards Analytical Laboratory was established. Concurrently, a formal Network of Analytical Laboratories was established from the Member States' *ad hoc* arrangements. At the IAEA's request, additional analytical laboratories were nominated by Member States. [2]

The primary purpose of the NWAL was to provide measurements for the verification of State-declared information, involving the comparison of the operator's and the NWAL laboratory's analytical results.

NWAL laboratories also became involved in the development of analytical techniques through Member State Support Programmes, providing support for the disposal of analytical residues, and the provision of reference material. The NWAL was also expected to act as a back-up to SAL in case of catastrophic failure, so that the safeguards analyses could continue to be completed in a timely fashion. [2]

Over the first ten years (1975-1985), the use of SAL and the NWAL increased from approximately 500 to 1500 samples per year. By 1985, the NWAL consisted of a total of 18 laboratories providing analytical services for the measurement of nuclear material, heavy water, and the production of reference materials. [2]

In the late 1980s and early 1990s there were a number of changes that affected the Agency's priorities. The number of laboratories that actively participated in NWAL began to decline, in part due to tightening of transport regulations for radioactive materials, and because of loss of member laboratories as their mission changed (e.g. laboratory management decided to no longer accept large quantities of nuclear material). In 1997, with the Agency's emphasis on strengthened safeguards, the Board of Governors was informed that "…among the new technical measures, priority has been given to the use of environmental sampling". Environmental sample analysis by the NWAL began with qualified laboratories in four Member States. [3]

By the year 2000, the number of NWAL laboratories performing environmental sample analysis had grown to 13 while the number of active laboratories conducting nuclear material analysis had dropped to seven. Further changes in the nuclear aspirations of Member States and commercialisation of laboratories further decreased the number of laboratories providing nuclear material analysis. Between the emphasis on the expansion of capacity and capability of providers of environmental sample analysis, and the attrition of nuclear material laboratories, by 2006 only one NWAL laboratory (aside from SAL) was actively providing analytical services for nuclear material analysis, but 13 laboratories (or groups of laboratories) in seven Member States and the European Commission were providing services for environmental sample analysis. [4]

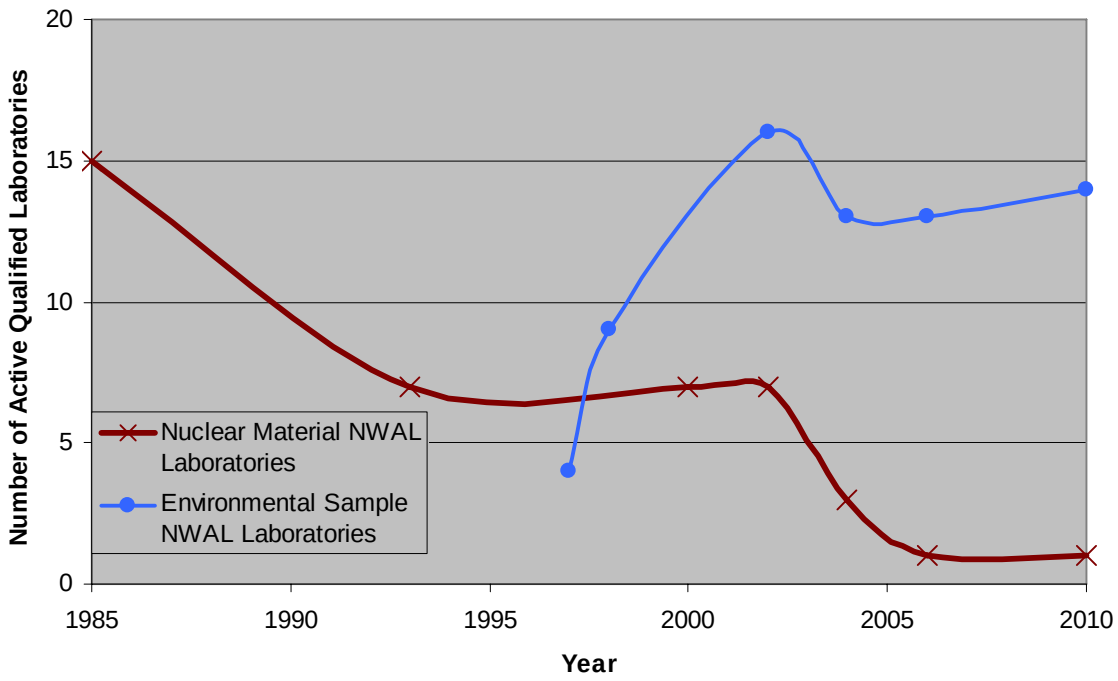

Figure 1. Graphic representation of growth and decline of NWAL providers for nuclear material analysis (dark red line) and environmental sample analysis (blue line). The connecting lines were drawn to illustrate the change between data points and are an estimate of that change. The NWAL members that supply reference materials and perform heavy water analysis are not represented on this figure.

3. Current Status of NWAL and its Requirements

As it currently stands, the NWAL provide support in four different categories: nuclear material, heavy water, environmental sampling and reference material. The sections below describe for each category: the contributing laboratories, sample capacity, and future requirements.

3.1 Environmental Sample Analysis in the NWAL

3.1.1 Current Status

Environmental samples are generally analyzed by two complementary measurement types: bulk and particle analysis. Bulk analysis is performed on the whole sample and determines the total amount of U and Pu as well as their average isotopic composition in the sample. Particle analysis techniques are used to measure the U and Pu isotopic composition of individual particles. The NWAL component for environmental sample analysis is a comparatively large and well-developed network, as shown in Figures 2 and 3.

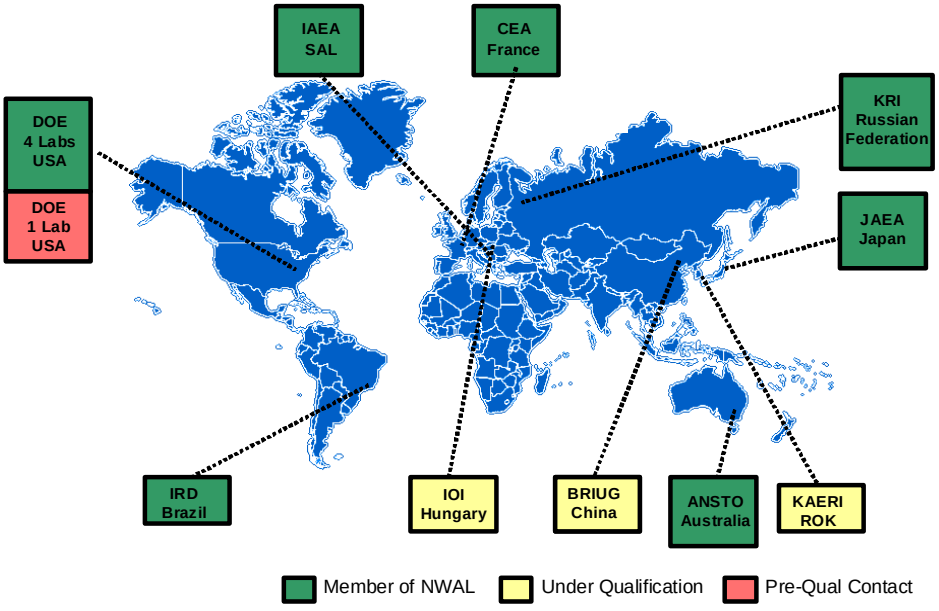

Figure 2. Map showing the distribution of member NWAL laboratories for bulk environmental sample analysis. Laboratories listed in green boxes are qualified, active members in NWAL. Laboratories listed in yellow boxes are laboratories that are currently in the process of qualifying for NWAL. Laboratories listed in the orange boxes have expressed interest in qualifying for NWAL, but have not yet begun the qualification process.

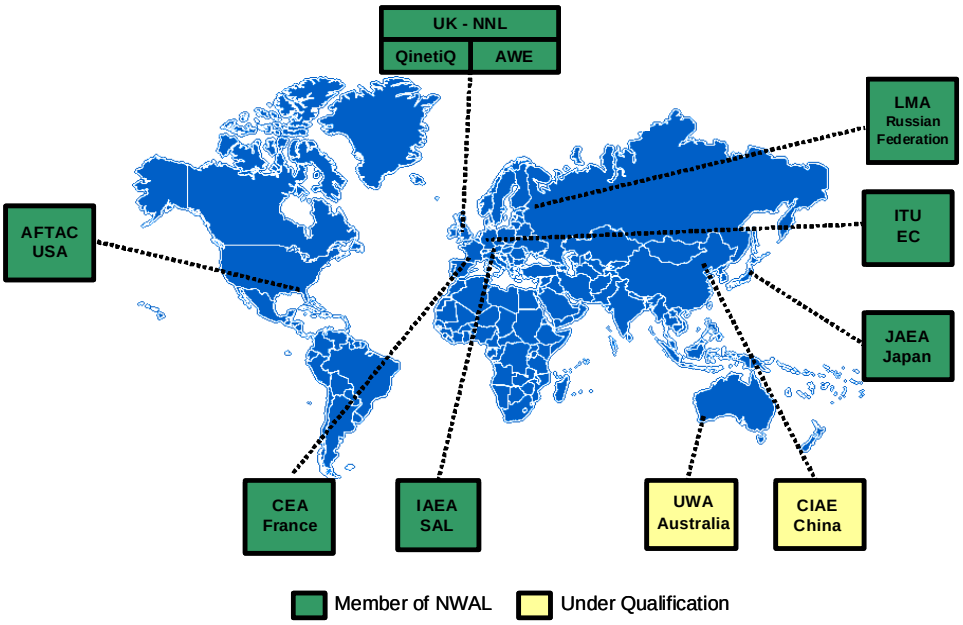

Figure 3. Map showing the distribution of member NWAL laboratories for particle environmental sample analysis. Symbols are the same as listed in Figure 2.

The IAEA is currently experiencing “bottlenecks” in sample analysis. Due to the ebb and flow of inspection samples, the IAEA experiences periods where there are more samples that need analysis than the NWAL has capacity to deal with. This results in delays for obtaining the analytical results.

The following two tables illustrate the current distribution of samples within the NWAL. In the last two years, approximately 35% of these samples have been determined to be high priority samples. High priority samples are not evenly distributed throughout the year and back up the queue for routine samples at NWAL facilities, thereby increasing delays for reporting results on the routine samples.

NWAL Member	**Total**
ANSTO (Australia)	10
CEA (France)	10
DOE (4 labs, US)	120
JAEA (Japan)	20
KRI (Russian Federation)	40
DOE (1 additional lab, US)	(Est. 40 in the future)
IOI (Hungary)	(Est. 30 in the future)
IRD (Brazil)	(Est. 40 in the future)
IAEA SAL	40
Total	**240**

Table 1. Utilized sample capacity for bulk environmental sample analysis within NWAL (some laboratories may not have been used to their full contracted capacity).

NWAL Member	SIMS	LG-SIMS	FT-TIMS	Total
AFTAC (US)	Upon request		400	**400**
UK NNL (AWE and QinetiQ)	60		10	**70**
CEA (France)	10		20	**30**
ITU (EC)	30	(est. 30 in future)		**30**
JAEA (Japan)	40			**40**
LMA (Russian Federation)	70			**70**
UWA (Australia)		(Est. 30 in future)		
IAEA-SAL	40 (0 in future)	(Est. 80 in future)		**40**
Totals	**250**	**(est. 140)**	**430**	**680**

Table 2. Utilized sample capacity for particle environmental sample analysis within NWAL (some laboratories may not have been used to their full contracted capacity).

3.1.2 IAEA Future Requirements

The IAEA is interested in expanding the capacity for particle analysis, in particular, for the determination of minor isotopes. The two most sensitive techniques for the measurement of minor isotopes are fission track thermal ionisation mass spectrometry (FT-TIMS) and large-geometry secondary ionisation mass spectrometry (LG-SIMS). These instruments and methods are expensive to purchase and maintain, and require a high level of technical expertise to use for best results. Qualification of a new instrument/technique under the best conditions, with previously trained technical experts can take years to complete.

The IAEA has identified a clear need to improve the timeliness of reporting results for both particle and bulk sample analysis by increasing the analytical capacities for environmental sampling. Aside from qualifying more laboratories, it would be beneficial if current NWAL members could expand their capacity. A number of NWAL laboratories provide analytical support to a very limited number of samples per year. Due to requirements for a minimum number of quality control (QC) samples, these laboratories end up analyzing a large fraction of QC samples compared to the number of Safeguards samples that the IAEA is able to send to them. For the most efficient QC to sample ratio, the IAEA recommends that NWAL laboratories consider a minimum capacity of 40 samples per year.

Another cause for the “bottleneck” is related to the very strict limits imposed by some laboratories on the levels of radioactivity that can be accepted at their facilities. Though this is not unexpected when dealing with the strict controls at environmental laboratories, the IAEA would benefit from increased capacity in laboratories that could accept “hot” environmental samples.

3.2 Nuclear Material Analysis in the NWAL

3.2.1 Current Status

Aside from SAL, for several years there was only one laboratory that was active in the NWAL for nuclear material analysis. In 2007, the Director General of the IAEA reported to the Board of Governors that “…the network for nuclear material analysis in addition to SAL consists of one fully active laboratory, which has a limited capacity for Agency samples. Consideration must be given to adding qualified laboratories to the NWAL capable of receiving and analysing nuclear material samples”. [5] Thus, the primary reason for expanding the network for nuclear material analysis is to provide QC for nuclear material analysis, and to act as a backup in case of emergency. If SAL is unable to perform safeguards analysis, there is only one laboratory that could take its place. In its turn, this facility is not necessarily ready to absorb 800 samples at a moment’s notice.

As shown in Figure 4, there are currently two qualified laboratories in the network and two laboratories that were qualified, but are not currently active. The laboratories that have not been active will need to analyze a series of QC standards before they can be re-activated.

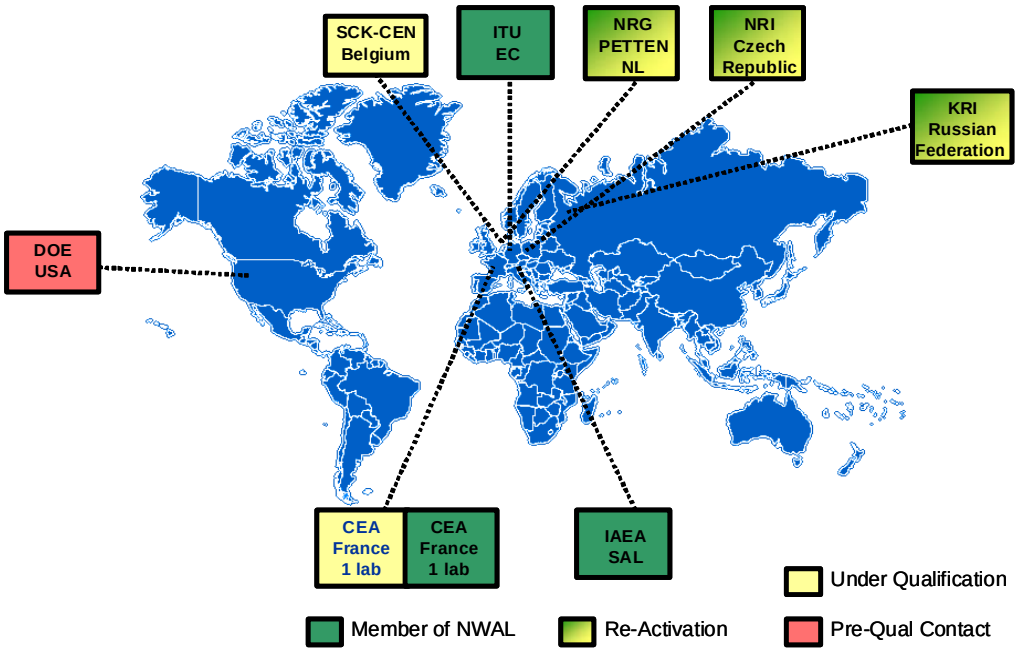

Figure 4. Map showing the distribution of NWAL laboratories for nuclear material analysis. The symbols are the same as in Figure 2, with the addition of the yellow and green shaded boxes. These represent laboratories that were qualified members of the NWAL but are currently not active.

Qualifying and re-activating laboratories that provide analytical services for nuclear material analysis is complicated due to the many types of samples and analysis methods that are used to analyse the samples. Table 3 illustrates the current nuclear material sample load for the IAEA-SAL.

Method	U Product	UF_6	U Various & Scrap	Spent Fuel, HALW	PNH, MOX (mg)	Pu (g)	Percent of Total
U Assay (D & G)	395	56	19				23%
U Assay (IDMS)			99	44	14		8%
U Isotopic (TIMS)	317	175	150	45	14		35%
Pu Assay (IDMS)				147	63	16	11%
Pu Isotopic (TIMS)				147	63	16	11%
^{238}Pu (Alpha Spec)				147	63	16	11%
Total Analyses	**712**	**231**	**268**	**531**	**221**	**48**	**2011**
Total Samples	**232**	**173**	**169**	**150**	**72**	**16**	**812**

Table 3. Number and Types of nuclear material samples analyzed at IAEA-SAL.

3.2.2 IAEA Future Requirements

The shipment of nuclear material samples to NWAL laboratories is seen as one of the primary sources of delays. Because of delays due to shipment of samples, the IAEA expects to continue to analyze the majority of the routine nuclear material samples at SAL. However, a number of samples will need to be distributed to the network to maintain a state of readiness for shipment and analysis of samples.

The IAEA will need to develop a robust QC programme to monitor the performance of each of the NWAL laboratories providing nuclear material analyses. Since this component of the network has not been active for a number of years, there is currently no QC programme specifically tailored to the needs of IAEA safeguards nuclear material analysis within the NWAL. This programme will also be complicated by the number of types of samples and analyses that may be requested for nuclear material safeguards samples (see Table 3). There will also be an increased need for QC materials, which will impact the reference material suppliers.

Further expansion of the network is under review but is not foreseen until the laboratories shown in Figure 4 have been qualified or re-activated, and the NWAL QC programme for safeguards nuclear material sample analysis is in place.

3.3 Heavy Water Analysis NWAL

3.3.1 Current Status

There is currently only one laboratory (KFKI Atomic Energy Research Institute – in Hungary) performing analysis on heavy water (D_2O). Similar to the situation for nuclear material analysis, this creates a single point of failure. If something happens at KFKI, that affects the laboratory's ability to perform the analyses, the Agency will be unable to obtain results for heavy water samples. Contacts have been made with a laboratory in Argentina that may be interested in qualifying to measure heavy water, and the IAEA is also considering whether heavy water analysis should be performed at SAL.

3.3.2 IAEA Future Requirements

The NWAL needs to have at least one additional laboratory that can provide QC and emergency back-up for the current NWAL laboratory providing heavy water analysis. A high priority need also exists for heavy water reference materials.

3.4 Reference and Quality Control Material Suppliers in the NWAL

3.4.1 Current Status

Figure 5 shows the distribution of laboratories that supply reference and QC materials to the NWAL. Though this component of the network is nearly complete, there is the possibility of adding one Canadian laboratory, which has offered to provide heavy water reference materials.

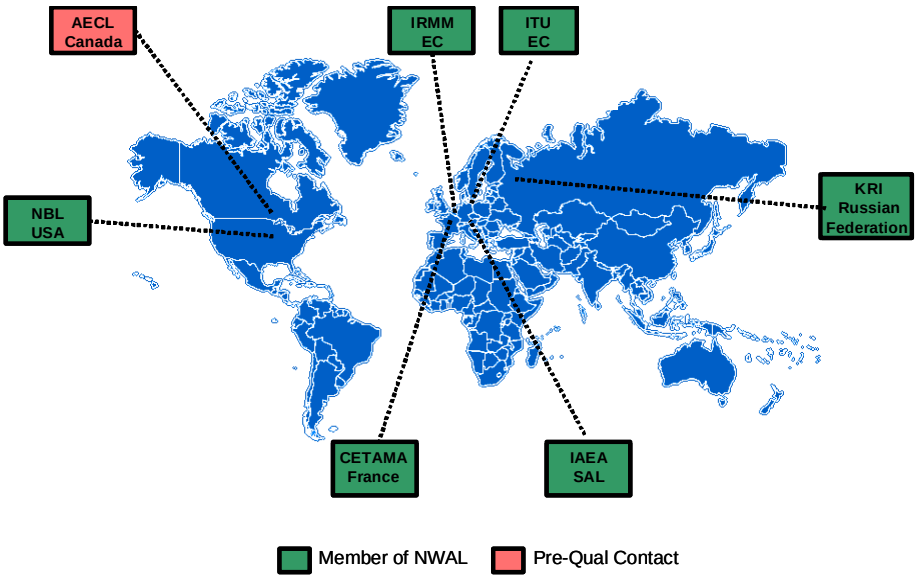

Figure 5. Map illustrating the distribution of NWAL laboratories that provide reference and quality control materials. Symbols are the same as in previous figures.

3.4.2 IAEA Future Requirements

As part of the QC programme for the NWAL, the IAEA encourages member laboratories to participate in inter-laboratory measurement exchange programmes. The IAEA also encourages the reference material and QC material providers to administer more of these round-robin measurement programmes, especially for environmental sample analysis.

The IAEA will continue to need reference and QC materials to be developed for bulk and particle environmental sample analysis. With the expansion of NWAL laboratories providing analytical services for nuclear material, the IAEA will have an increasing need for appropriate QC test samples and standards. In addition, the IAEA needs relevant reference materials for special analysis, such as trace element determination.

The IAEA will also continue to coordinate with reference material and QC material suppliers to determine which laboratory will provide what reference material to the safeguards and nuclear community.

4. Recommendations

After completing this historical review of the NWAL, a number of recommendations for improvement and continued maintenance became apparent. They are presented below for consideration.

To reduce “bottlenecks”, increasing the capacity of NWAL laboratories (number of samples analyzed per year) for both bulk and particle environmental sample analysis would be more efficient and effective than adding more laboratories in the NWAL..

The distribution of samples will continue to be reviewed in order to reduce the bottlenecks, and to use the laboratories as efficiently as possible, including: looking at sample batch size (what is too big or too small?), obtaining a “green light” notification from laboratories regarding ability to accept samples, especially for those with smaller capacities; and reducing the number of high priority samples.

Presently, the IAEA has embarked on an ambitious programme for qualifying additional laboratories. The IAEA is currently in the process of qualifying four nuclear material laboratories, one laboratory for analysis of particle environmental samples, three for bulk environmental sample analysis, and possibly one for heavy water analysis. As the qualification process is time-consuming and work -intensive for both the IAEA and for the laboratory that is trying to qualify, in the short term, the IAEA does not intend to significantly expand the network beyond these new laboratories.

Since the beginning of NWAL, candidates for NWAL have been required to have developed and implemented a quality assurance (QA) system. The IAEA’s qualification process for NWAL laboratories currently relies significantly on elements of ISO 17025. Some NWAL members are fully ISO 17025 compliant, but this is not a requirement for participation in NWAL. Other NWAL members, as well as future candidates, may wish to consider full ISO 17025 compliance.

The Agency must remain an unbiased and independent organisation. The IAEA Director General has identified the issue of the Agency maintaining a high degree of analytical capability. It is therefore important to maintain the IAEA’s own laboratories (SAL) and the NWAL as a Centres of Excellence for safeguards measurements.

5. Conclusion

Since the NWAL was implemented, the IAEA has also modified and strengthened its safeguards programme. The composition and capabilities of NWAL has changed over the past 36 years in order to meet IAEA safeguards analytical needs. During this period, in particular while NWAL was striving to develop and implement new capabilities, support for some analytical capabilities may have been lost. To address the potential shortcomings in analytical capabilities, the IAEA is striving to re-establish a balance of capabilities for nuclear material, environmental sample analysis and heavy water safeguards analysis within the NWAL and SAL. Finally, the IAEA’s NWAL is a unique and mutually beneficial initiative, and the IAEA appreciates the support of Member States and the European Commission and for their continued participation and development of this programme.

6. References

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Recent developments for COMPUCEA 2nd generation

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Abstract:

*COMPUCEA (**Com**bined **P**rocedure for **U**ranium **C**oncentration and **E**nrichment **A**ssay) is used for on-site analytical measurements in support of joint Euratom-IAEA inspections during physical inventory verification (PIV) campaigns in European Low-Enriched Uranium (LEU) fuel fabrication plants. The analyses provided on site during the PIV involve the accurate determination of the uranium element content and of the ^{235}U enrichment in verification samples (uranium product samples of solid form , i.e. powders, pellets) selected by the Safeguards inspectors. These samples are dissolved and then measured by energy-dispersive X-ray absorption edge spectrometry (L-edge densitometry) to obtain the uranium elemental concentration and gamma spectrometry with a Lanthanum-bromide detector for the ^{235}U abundance determination. The second generation of COMPUCEA equipment is compact, rugged and ready-to use directly after transport. A new set of reference materials has recently been obtained. Examples from measurement campaigns and round robin exercises that demonstrate the performance of the technique are shown. The use of COMPUCEA for IAEA purposes has successfully been demonstrated during a campaign outside EU. The use of a Monte Carlo routine developed in-house to calculate the enrichment correction factor, accounting for small variations of relevant sample parameters, has been validated by comparison with data from primary reference methods.*
For the determination of the ^{235}U enrichment, knowledge of a possible neutron absorber content (mainly Gd, added to the fuel as burnable poison) in the sample is required to accurately calculate self-absorption effects. A new independent unit for X-ray fluorescence measurements was designed for the Gd-pre-screening of the samples. First data obtained for solid and liquid samples are discussed.

Keywords: uranium concentration assay, uranium enrichment, LEU fuel fabrication, Monte Carlo simulation

1. Introduction

COMPUCEA (**Com**bined **P**rocedure for **U**ranium **C**oncentration and **E**nrichment **A**ssay) is used for analytical measurements in support of Safeguards inspections during accountancy verification campaigns in Low-Enriched Uranium (LEU) fuel fabrication plants. The analyses are provided directly on site with transportable equipment. They involve the accurate determination of the uranium elemental content (reported as mass fraction) and the ^{235}U enrichment in samples (uranium oxide pellets and powders) selected by the Safeguards inspectors.

The main advantage of the technique is that high precision analytical results are quickly and directly reported to the inspectors on-site, thus ensuring timeliness and at the same time eliminating the necessity to ship radioactive samples to a Safeguards laboratory.

2. Analytical procedure

The complete COMPUCEA analysis procedure represents a combined chemistry-spectrometry analysis involving accurate analytical steps (like quantitative sample dissolution, solution density measurements, quantitative aliquoting, etc.) combined with radiometric measurements. The radiometric techniques involved are X-ray absorption edge spectrometry at the L_{III} absorption edge of uranium (L-edge densitometry) and passive gamma counting with a $LaBr_3$(Ce) detector. The techniques are briefly described below: more detail can be found in [1,2]. The general scheme of analysis includes the following main steps:

- ***Sample preparation:*** The first step is to transform the solid uranium samples (powders or pellets) into a uranyl nitrate solution of approximately constant acidity (3 M) and uranium concentration level (ca. 190 gU/L), which is then characterised for its density and temperature. The analytical tools needed for this sample preparation step (hot plate, density measurement device, glassware, pipettes etc) are brought on site as part of the COMPUCEA equipment, but the use of operator facilities (fume hood, analytical balance) is also required at this stage.
- ***Radiometric measurements (L-Edge Densitometry and Gamma Spectrometry):*** Aliquots are taken from the sample solution and subjected, without any further treatment, to parallel L-edge densitometry and passive gamma counting. Prior to the measurement campaigns, the equipment is pre-calibrated at ITU and then calibrated again on-site using certified reference material (sintered UO_2 pellets), stored at each facility under common Euratom/IAEA seal.
- ***Data evaluation:*** A user-friendly software package for instrument control and data handling is utilised. In the final step of evaluation, the data obtained from the sample preparation and from the two radiometric measurements are combined to evaluate the uranium weight fraction in the original sample and the ^{235}U weight fraction in the uranium material. The two radiometric measurements are interdependent, i.e. each technique requires input from the other for the final data evaluation.

2.1. Measurement techniques

Uranium concentration determination by L-edge densitometry

This technique uses the element-specific discontinuity of the photon absorption that occurs when the photon energy exceeds the binding energy of the electrons in the U L-shell (L-edge absorption spectrometry). The setup for uranium elemental assay consists of a miniaturised X-ray source and a high-resolution Peltier-cooled silicon drift detector. The X-ray beam is collimated and passes through a quartz cuvette of well-defined path length, which contains the sample solution. The cuvette used here is a 2 mm flow-through cuvette, into which the sample is loaded using a syringe. After each measurement, the cuvette is rinsed and dried before loading the next sample into the same cuvette. The calibration is performed with the same cuvette. This overcomes the issue of any variation in path length between manufactured cuvettes, which would otherwise add to the overall uncertainty of the measurement. The transmitted spectrum is recorded by the silicon detector as a function of X-ray energy, showing a characteristic jump of the photon transmission at the L-absorption edges of uranium. The height of this jump is proportional to the uranium concentration in the solution. The uranium concentration is evaluated from the spectral data around the L_{III} absorption edge, which offers the largest differential change in the photon attenuation. The procedure for data evaluation follows the proven procedure applied with the K-edge densitometry technique (ISO 13464:1998) [3]. The initial result obtained is the concentration of uranium in solution in g/L. The density of the solution (in g/cm^3) is then used to convert from g/L into g/g. The uranium elemental content as mass fraction of the original sample is then calculated from the solution and sample weights. The enrichment (obtained from the gamma measurement) is needed to obtain the correct average atomic weight.

^{235}U enrichment determination with a $LaBr_3$(Ce) detector

The ^{235}U enrichment measurement in the 2nd generation of COMPUCEA is based on the counting of the ^{235}U 186 keV photons of a defined amount of uranium in solution in a well-defined counting

geometry. The detector used is a standard-type 2" x 1" cerium-doped lanthanum bromide scintillation detector – $LaBr_3(Ce)$. A significant advantage of this detector for in-field use is that it operates at room temperature and, therefore, there is no requirement for an on-site supply of liquid nitrogen for detector cooling. Furthermore, the detector is ready for use immediately after unpacking of the equipment.

The relatively simple gamma spectrum of ^{235}U allows accurate enrichment measurements at the lower energy resolution of the $LaBr_3$ compared to High-Purity Germanium (HPGe) detectors (FWHM @ 186 keV of approximately 9 keV for the LaBr detector compared to a value of 1.3 keV obtained with the HPGe well detector used previously).
A two-step process is used to evaluate the gamma spectrum in order to obtain an accurate enrichment value: (1) analysis of the gamma spectrum itself [4] for the extraction of the 185.7 keV net peak counts, and (2) calculation of appropriate correction factors for the extracted peak counts, accounting for the impact of relevant sample parameters including: concentration of uranium in the solution (obtained from the parallel L-edge densitometry); solution density; presence of neutron absorbers such as Gd; and bottom thickness of the sample container (which has an influence on the measurement geometry). These (small) corrections are calculated relative to a standard configuration either by using separate correction factors or by modelling the measurement configuration by a Monte Carlo simulation (both options have been implemented, a comparison with data from primary reference methods for validation is shown in Section 3.1.2.). The final result is the ^{235}U abundance in units [wt% ^{235}U/U].

2.2. Reference materials

The reference materials available for calibration consist of a set of sintered UO_2 pellets with different enrichment grades. The UO_2 pellets were taken from the production batches of a uranium fuel fabrication plant, and subsequently characterized by primary analytical methods for the uranium element content and isotopic composition. The analytical measurements for material characterisation were independently carried out by ITU and IAEA-SAL. A set of such pellets with nominal enrichments of 0.72%, 2% and 4% ^{235}U has been available for several years, a new set of 100 pellets with 3% ^{235}U and 100 pellets of 4.4% ^{235}U has recently been purchased. The joint characterisation by ITU and IAEA-SAL is ongoing, each of the labs verifies the uranium elemental content (by titration) and ^{235}U-enrichment (by Thermal Ionization Mass Spectrometry, TIMS) independently.
In addition, 200 certified reference pellets (CRM 125-A), obtained from New Brunswick Laboratory (NBL), USA, are available.

In order to ensure continuity of knowledge of the pellets, a common Euratom/IAEA inspection was organized during which samples were taken to certify the different batches. The remaining pellets were split for use at the different European installations and ITU and placed under Common Seals. Each of the packages contains 3 types of pellets: pellets of 3% and 4.4% enrichment and NBL pellets (kept in their original container, each pellet is separately packaged). All samples for the same location were placed inside a Biopack2 –Type A container.

The reference solutions required for instrument calibration, either in field or at ITU, are prepared from these reference pellets, following exactly the same procedures for sample preparation as applied for the normal measurement samples. From each reference pellet, a single reference solution is prepared, which is used both for the calibration of the L-edge densitometer and of the gamma spectrometer.

3. COMPUCEA performance

To evaluate the performance of the COMPUCEA 2nd generation, the influence of relevant measurement parameters was studied in detail and an estimate of the total uncertainty of the two analytical determinations was made, which includes uncertainty components related to the sample preparation [1].
The estimations for the total measurement uncertainties of COMPUCEA 2nd generation are well within the International Target Values (ITV) for measurement uncertainties in the field of International Safeguards for nuclear materials (1sigma relative combined standard uncertainty: 0.28% for U concentration and 0.45% for ^{235}U abundance) [5].

For method validation, the measurement performance has been evaluated by comparing the COMPUCEA results

- with results from parallel analyses made with a primary reference method,
- with well-specified reference values for the quantity of interest, and
- with data obtained in round robin tests

Results of these studies have been reported in [1,2,6].

3.1. Comparison of COMPUCEA and primary reference methods

For validation of the technique, COMPUCEA results have extensively been compared to analyses obtained using qualified primary analytical methods (typically potentiometric titration according to the method of Davies and Gray for uranium concentration and Thermal Ionisation Mass Spectrometry, TIMS, for uranium enrichment determination) at ITU and elsewhere. This comparison is continuously updated, both for in-field samples and samples analysed at ITU.

3.1.1. Infield performance / demonstration exercise for IAEA outside EU

For in-field measurements, COMPUCEA data are typically compared to results obtained from a sub-set of samples taken in parallel and shipped for remote analysis (by IAEA-SAL). For all samples measured with COMPUCEA 2nd generation in 2007, 2008 and 2009 there is excellent agreement with the data from IAEA-SAL, well within the uncertainty levels set by the ITVs. For 2010 no data for comparison are available yet. More detail can be found in [6].

As part of the Euratom Support Program to the IAEA, a COMPUCEA demonstration exercise in a LEU fuel fabrication plant outside Europe was performed in October 2010 in Kazakhstan, jointly with IAEA inspectors, to evaluate the use of COMPUCEA for Safeguards purposes outside the EU. The goal of the visit was to reproduce the real working conditions of the in-the-field COMPUCEA method during a Physical Inventory Verification (PIV), as a contribution to the authorisation process for COMPUCEA as IAEA Safeguards equipment. For the purpose of the exercise, all measurements were made using the archive samples of the PIV 2009 in order to avoid a new sampling campaign. For calibration, one of the archive pellet samples was selected, to avoid the time-consuming shipping of certified reference material. The measurement results were then compared to the IAEA laboratory results, to assess the method performance at the end of the exercise.

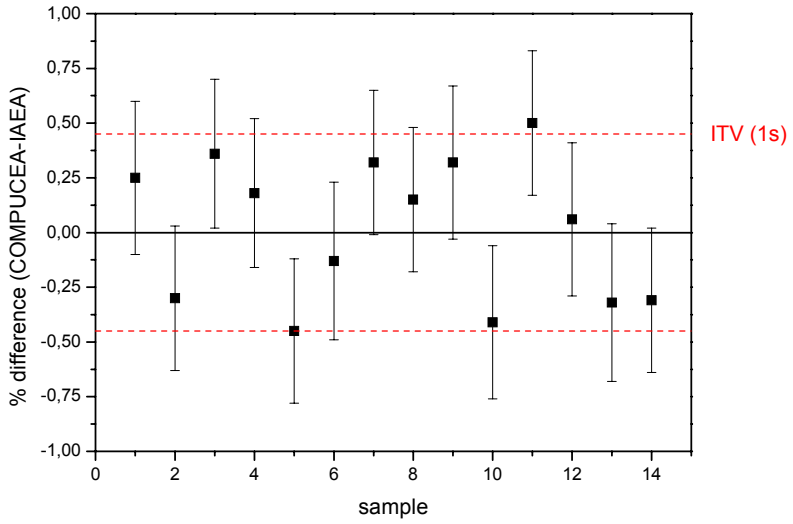

Figure 1: Percentage difference between ^{235}U enrichment measured with COMPUCEA 2nd generation on-site at a fuel fabrication facility in Kazakhstan and data obtained by IAEA-SAL for samples taken in parallel. The error bars are 1s combined uncertainty (including the uncertainty of the SAL result for the pellet used for calibration).

To demonstrate the potential capacity of the method, 15 out of the 20 samples taken during PIV (+ 1 pellet for calibration) were analyzed during 5 working days (including setup and re-packing of the equipment and calibration). The selected samples were considered as most representative of the type of material present at the facility. The selection included pellets, clean powder and scrap material (clean and dirty). The analysis of all 16 samples within 5 working days including packing and unpacking proved the capacity of the method to analyze the entire batch of samples collected during the duration of a typical PIV, including the evaluation of the results and intermediate report drafting.

The analytical performance was highly satisfactory. The results regarding enrichment were in good agreement with IAEA laboratory results, as can be seen in Figure 1. An average difference between COMPUCEA and IAEA-SAL of 0.02% with a standard deviation of 0.32% can be calculated, which is well within ITV. The uncertainties are slightly higher than for other COMPUCEA campaigns, due to fact that during this exercise an archive pellet sample was used for instrument calibration, with associated higher uncertainty, rather than a certified standard.
On powder samples, the uranium concentration results reflected a moisture effect during the sample storage which is the consequence of analyzing archive samples after more than 12 months. For pellet samples, which suffer less from moisture effects, the uranium concentration results were in perfect agreement.

3.1.2. Comparison of COMPUCEA and TIMS - Validation of Monte Carlo calculation for enrichment correction factor

For ^{235}U enrichment determination, the COMPUCEA data evaluation involves a two-step process: (1) analysis of the gamma spectrum itself for the extraction of the 185.7 keV net peak counts, and (2) the calculation of appropriate correction factors for the extracted peak counts accounting for the impact of variable sample parameters.
The correction factors in step (2) are calculated relative to a standard configuration. The main contributors (which are typically below 0.5%) are the following:

- Variation of the concentration of the uranium solution (standard concentration: 190 gU/l, normal concentration range: 175-195 gU/l). Here, a correction for the self-attenuation effect of uranium needs to be made for variations of the uranium concentration around the adopted standard concentration. The results obtained by L-edge densitometry are used for the calculation.
- Gd presence in the solution, which leads to self-attenuation of the sample depending on the Gd concentration (standard configuration: no Gd). The estimation of the Gd concentration is currently obtained from the L-edge densitometry measurement (comparison of "extrapolated fitting" vs. "non-extrapolated fitting" result, see [1] and Section 4.).
- Container bottom thickness (standard configuration: 1.10 mm). The bottom thickness of the sample containers represents a crucial parameter, because even small variations slightly change the sample-to-detector distance and hence also the measured 186 keV count rate. Prior to the in-field campaigns, the bottom thickness of each container is measured with a thickness gauge with an accuracy of $\pm$ 0.01 mm. For each individual campaign, a set of containers is selected which shows little variation in bottom thickness.
- Interfering gamma rays, i.e. the daughter products ^{234}Pa and ^{234m}Pa from ^{238}U.

The corresponding correction factors can be obtained in 2 different ways: using separate correction factors for each of the major contributors according to data obtained from experimental studies (= "traditional approach") and (ii) by a Monte Carlo (MC) calculation routine, which calculates the detection probability for the 185.7 keV photons for the given measurement configuration in dependence of the relevant sample parameters uranium concentration, Gd concentration and container bottom thickness relative to the standard configuration. The Monte Carlo calculation routine has been developed in-house. The relative detection rates calculated within a runtime of 100 s for the Monte Carlo calculation have a statistical precision of 0.03%.
The correction factor for interfering gamma rays (the daughter products ^{234}Pa and ^{234m}Pa from ^{238}U) is applied for both approaches.

A comparison of the ^{235}U enrichment correction factors with the two different methods was done using measurement data from in-field campaigns. This study is presented in [6]. The data show good agreement within the statistical uncertainty of the Monte Carlo calculation. However, for the samples

studied, the parameters (especially uranium concentration in solution) were very close to the standard configuration.

A comparison between COMPUCEA and TIMS results for ^{235}U enrichment, which covers a wider variation of sample conditions (uranium concentration in solution and Gd content in the sample), is presented here, using experimental data from a series of 63 uranium oxide powder and pellet samples. These samples from fuel fabrication plants were analysed at ITU, both by primary reference methods (titration and TIMS) and COMPUCEA. The comparison between COMPUCEA and TIMS for ^{235}U enrichment in samples without Gd is presented in figure 2, as a function of uranium concentration in the solution. The normal operation range is indicated by the green shaded area. The error bars shown are 1s combined uncertainty (COMPUCEA and TIMS). The COMPUCEA results were obtained using the 2 different methods for enrichment correction factor calculation described above. Data shown in blue have been obtained using the traditional approach; data in purple have been obtained using the Monte Carlo calculation tool. The zero-line (black straight line) is shown, together with the average difference (straight line, blue/purple; average for traditional calculation: -0.02%, Monte Carlo calculation: -0.08%) plus/minus standard deviation (dashed line, blue/purple; standard deviation for traditional calculation: 0.23%, Monte Carlo calculation: 0.18%).

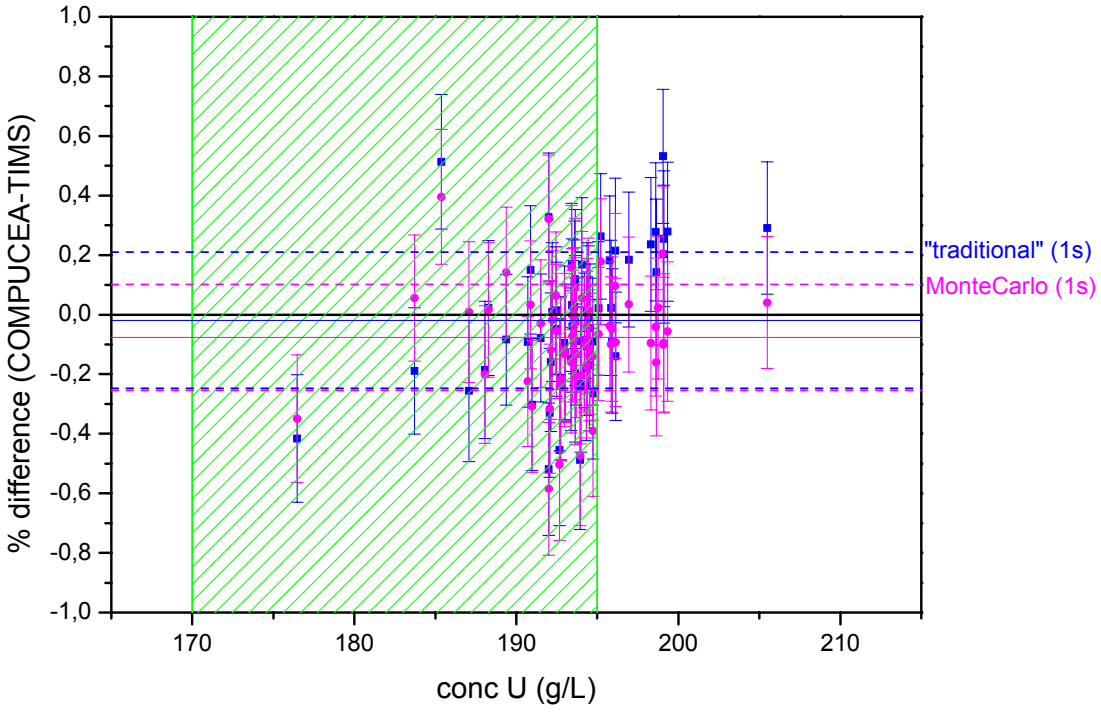

Figure 2: Percentage difference between ^{235}U enrichment values measured with COMPUCEA 2nd generation vs TIMS. A detailed explanation is given in the text above.

In conclusion, it can be said that all results are well within ITV and show no significant bias. The results obtained using the Monte Carlo calculation tool show slightly smaller overall variation (i.e. standard deviation), especially in cases which show stronger deviation form the normal operating range (as can clearly be seen in the graph for uranium concentrations above 195 g/l).
8 samples containing Gd were evaluated separately. Here, the average difference calculates to 0.38% with a standard deviation of 0.24% for data obtained using the traditional calculation and 0.15% with a standard deviation of 0.21% for Monte Carlo calculation. The results obtained with the Monte Carlo calculation are clearly in better agreement with the TIMS values, both for accuracy and precision.
For future campaigns, the Monte Carlo tool will become the standard routine used during the data analysis procedure, it is currently being included in the analysis software package.

3.2. Participation in round robin tests

COMPUCEA regularly takes part in round robin exercises. During 2010, COMPUCEA participated in a round robin exercise offered by New Brunswick Laboratory (NBL), USA, under the Measurement Evaluation (SME) program, where uranium samples (UO_2 pellets, U_3O_8 Powder, UF_6 samples) are distributed for destructive analysis. UO_2 pellet samples were analysed by COMPUCEA during several rounds of SME.

The results of 3 rounds, where reference data is already available, are shown in Figure 3. The COMPUCEA data are in good agreement with the reference values (average difference: 0.04% (straight blue line) with standard deviation 0.06% (dashed blue lines) for uranium concentration, average difference: -0.07% (straight blue line) with standard deviation 0.19% (dashed blue lines) for enrichment - the reported data were obtained using the traditional approach for calculating the enrichment correction factors, as the Monte Carlo tool had not yet been implemented) and well within the ITVs (ITV (1s) dashed red line, ITV (2s) straight red line).

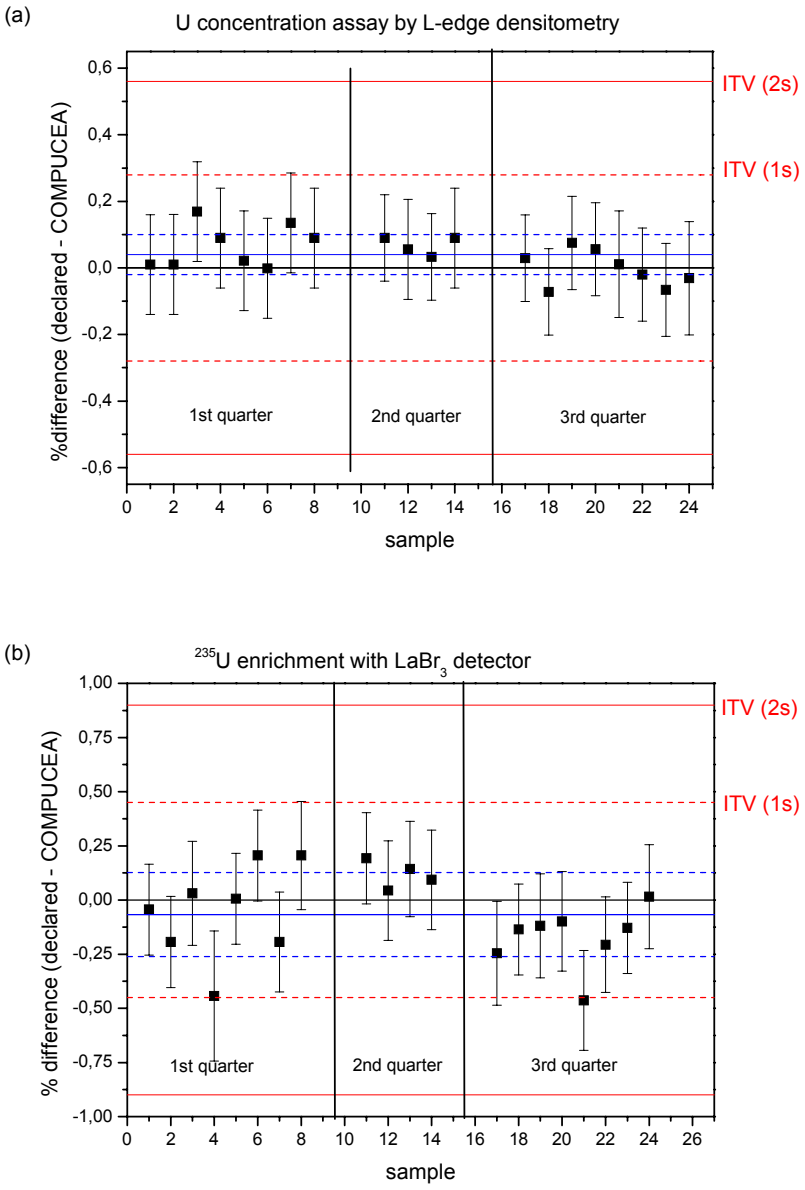

Figure 3: Percentage difference for (a) uranium concentration and (b) ^{235}U enrichment values measured with COMPUCEA vs. the declared values for the SME program. More information can be found in the text.

4. Recent developments: XRF unit

For the final evaluation of the ^{235}U enrichment, it is necessary to determine the Gd content in the sample to accurately calculate the respective correction factor. A first value can be obtained from the L-edge data by using the extrapolated and the non-extrapolated results [1]. However, this is only an indirect measure, as a difference in the 2 values only points to a general difference in matrix composition relative to the standard conditions, but no direct measurement of Gd itself is made. The latter can be achieved, for example, by an additional XRF measurement. First tests were done using a modified sample holder in the L-edge setup. Here, the measurement geometry required the covering of the sample with a thin foil to avoid spilling of the liquid. This led to problems with absorption effects and unsatisfactory reproducibility of the results.

Thus, a new independent unit for XRF measurements was designed, using the same type of X-ray tube and Si drift detector as for the L-edge setup. These components are typically taken in-field as spare parts, so any additional equipment is limited to the small aluminium holder that fits the X-ray tube, detector and sample container. The XRF setup is schematically shown in figure 4. The setup was designed in such a way that a sample container with a liquid sample can be positioned in an upright geometry. This has the advantage that the container can be left open during the measurement, minimizing absorption effects in the X-ray beam paths (only air between X-ray tube, sample and detector).

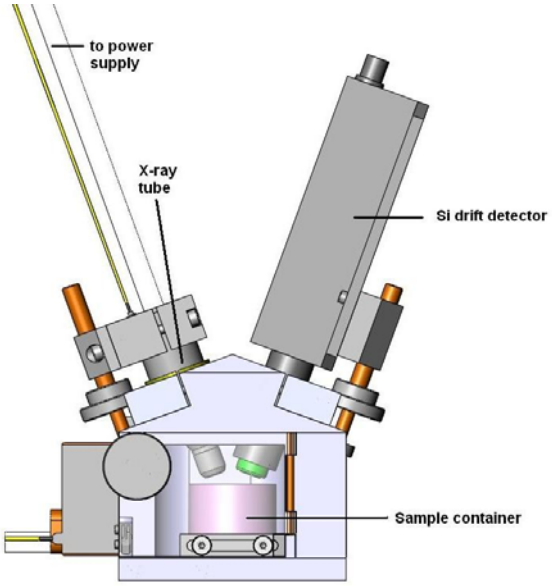

Figure 4: Setup for XRF.

The setup also offers the possibility to perform XRF measurements directly on solid samples. A set of Gd-containing UO_2 pellets with different Gd concentration (2%, 4%, 6% and 8%) were measured. The spectra were evaluated using Genie2000 to determine the peak areas for the main lines ($L_{\alpha 1}$ peaks) of Gd (6.06 keV) and U (13.61 keV), respectively. The ratio Gd/U showed a linear behaviour with increasing Gd content, so a calibration can be performed, which provides first information on the Gd content in pellet samples. It only serves as a rough estimate; variations in the measurement geometry strongly affect the Gd/U ratio here. This was tested by placing the same pellet in different geometries (up to 30% variation of the Gd/U ratio was observed). For placing the pellet in the same geometry but turning or moving it so different areas of the pellet surface would be irradiated by the X-ray beam, resulted in several % variation of the Gd/U ratio (relative standard deviation (1s) 2-8%, depending on the Gd content, higher uncertainties were obtained for low amounts of Gd).

More accurate results are expected from the use of solutions, as they are homogeneous. The data presented here are first results; a more complete study is still ongoing. A sub-set of the Gd-containing solutions used for the investigations presented in section 3.1.2 was selected. For data evaluation, Genie 2000 was used. In the future, different approaches (including software specifically dedicated to the analysis of XRF spectra) for extracting the net peak counts from the spectrum will be tested and compared to find an optimum algorithm which offers easy handling and good precision.

The repeatability was studied by measuring the same solution over an extended period of time, the samples were removed and stored in-between the individual measurements. The results for the experimentally obtained, uncorrected Gd/U ratio from a uranium solution with 8% Gd are shown in Figure 5, the error bars plotted are 1s, calculated as the standard deviation of several – typically 3 to 5 – measurement repetitions. A standard deviation of 0.7% can be calculated for the values covering a period of 2 ½ months.

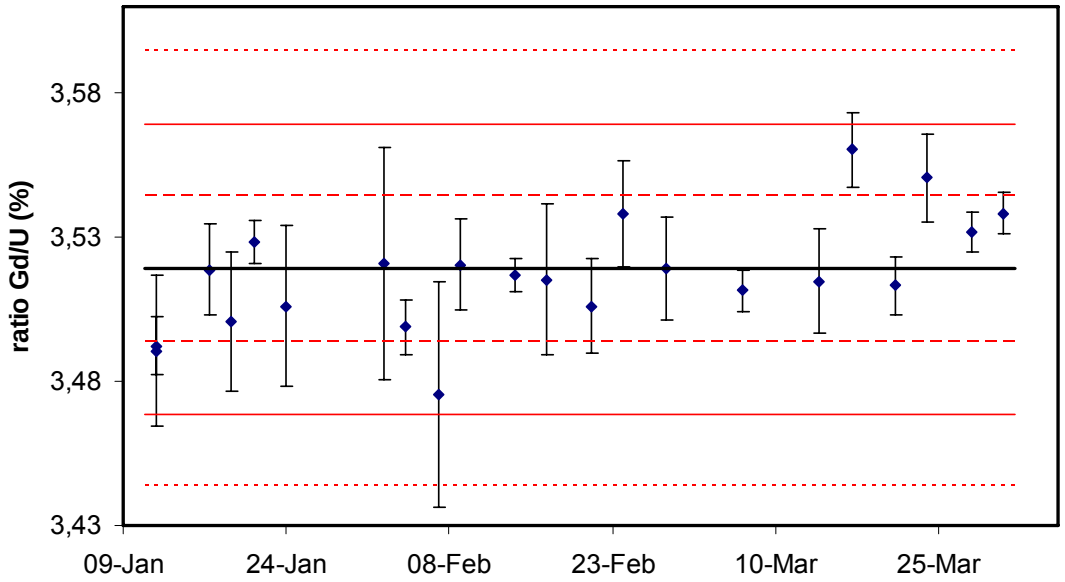

Figure 5: Repeatability of XRF measurements (experimental uncorrected peak ratio Gd/U) for a uranium solution containing 8% Gd (black solid line: average, red lines: standard deviation - dashed: 1s, solid: 2s, short dashed: 3s).

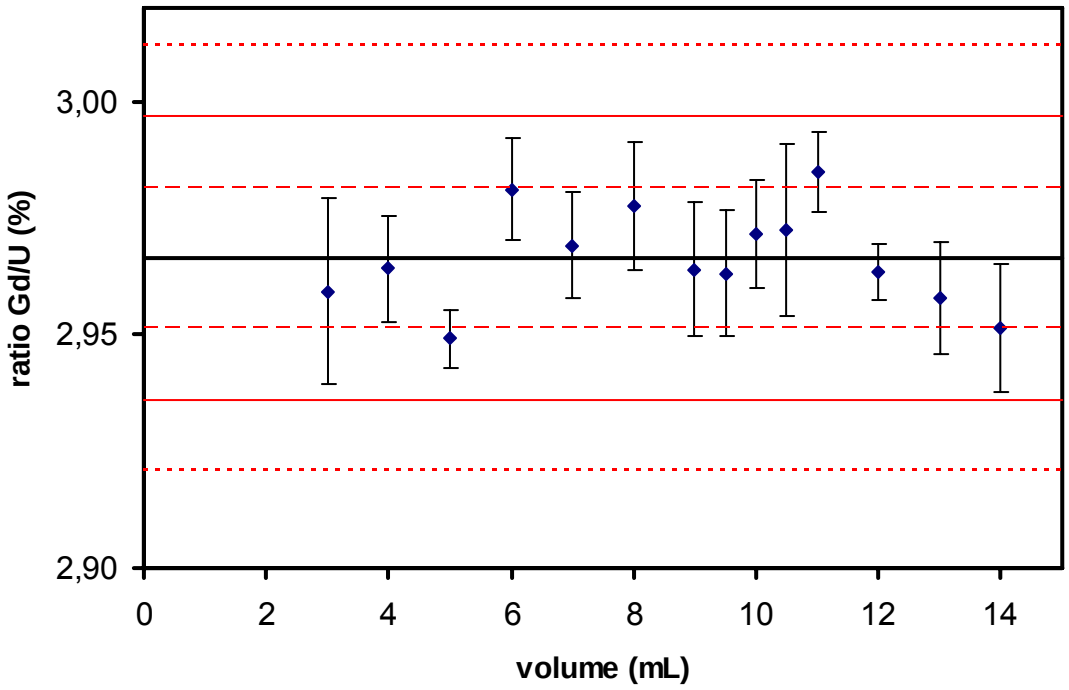

Figure 6: Effect of varying the volume of solution for XRF measurements (experimental uncorrected peak ratio Gd/U) of a solution containing 7% Gd (black solid line: average, red lines: standard deviation - dashed: 1s, solid: 2s, short dashed: 3s).

The variation of the U/Gd ratio was further studied as a function of sample height by varying the volume amount of solution in the container. The same sample containers as for enrichment measurements were used, without the lid. As the penetration depth of the low-energy L-X-rays in the solution is very low, only the surface and the first few hundred µm of the solution are relevant for the analysis. Therefore, only a small volume of solution is necessary and no big change in the U/Gd ratio is expected when increasing the volume of solution. This is indeed the case, as can be seen from Figure 6 (for a solution with 7% Gd). Over a wide range of varying the solution volume, the Gd/U ratio remained constant within the uncertainty of the measurement (the error bars plotted are 1s, calculated as the standard deviation of several – typically 3 to 5 – measurement repetitions, the overall standard deviation is 0.5%).

5. Conclusion

COMPUCEA is a compact and transportable system which allows high-accuracy uranium elemental assay and enrichment determination from solid uranium samples. The 2nd generation COMPUCEA avoids radioactive sample transport, does not need transport of radioactive sources and attains excellent accuracy with an easily transportable system. It is routinely applied in physical inventory verification campaigns at European LEU fuel fabrications plants; recently the performance has been successfully demonstrated in a campaign outside EU, jointly with IAEA. The performance is well within the International Target Values and is regularly monitored by comparison with primary reference methods and participation in round-robin exercises. A new Monte Carlo routine has been developed in house for calculation of enrichment correction factors, accounting for variations of relevant sample parameters, it has been validated by comparison with primary reference measurements. First studies have been performed with a new independent X-ray unit for Gd-pre-screening of samples.

6. Acknowledgements

The authors would like to acknowledge the invaluable support provided by Dr. Evelyn Zuleger and the Analytical Service group at ITU for the many measurements performed with primary reference methods for the validation of the techniques.

We wish to express our special thanks to New Brunswick Laboratory for supplying certified reference pellets under the Joint Action Sheet between Euratom and the U.S. Department of Energy.

7 Legal matters

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Determining the plutonium isotope ratio using alpha spectrometry

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Abstract:

Radiochemically processed sources containing isotopes of Pu were measured by alpha spectrometry. Alpha-particle energy spectra were also generated by simulations. The spectra were unfolded using a novel analysis program ADAM to determine the $^{239}Pu/^{240}Pu$ isotopic ratios and to test the uncertainty computation. It is shown that ADAM gives correct peak areas and uncertainty limits. The results indicate that alpha spectrometry alone can be reliably used for Pu isotopic ratio determination.

Keywords: alpha spectrometry; plutonium; isotopic ratio

1. Introduction

Isotopes of uranium and transuranium elements are of special concern in nuclear safeguards and non-proliferation. Alpha spectrometry and mass spectrometry are widely used methods to determine the isotopic composition. Unfortunately, in mass spectrometry the isobaric interferences between ^{238}U and ^{238}Pu hinder reliable determination of ^{238}Pu. In alpha spectrometry, the small energy difference between the main peaks of ^{239}Pu and ^{240}Pu (approximately 12 keV) makes their unequivocal determination extremely challenging.

A novel analysis tool known as ADAM (Advanced Deconvolution of Alpha Multiplets) has been recently developed to unfold complex alpha spectra [1-2]. The program can be applied to spectra obtained from different types of samples, such as air filters, swipes or liquid residues.

In the present study, we investigate the possibility of determining the $^{239}Pu/^{240}Pu$ isotopic ratios from radiochemically processed sources that also contain ^{238}Pu and ^{242}Pu. Results obtained by alpha spectrometry are compared to the reference values. In addition, a large number of simulated spectra was produced and analyzed by ADAM in order to show the validity of the uncertainty calculation.

2. Sources and measurements

A set of sources containing isotopes of Pu were prepared by NIST and analyzed by STUK as blind samples, i.e. the nuclide-specific activities were not known beforehand [3]. The amounts of ^{238}Pu and ^{242}Pu were equal in all sources whereas those of ^{239}Pu and ^{240}Pu varied. Here only the $^{239}Pu/^{240}Pu$ isotopic ratios are reported. The nuclides were processed to stainless steel disks and measured at source-detector distance of 48.5 mm. This distance ensures that the effect of the coincidences between alpha particles and low-energy photons/electrons is negligible [4].

The spectra were recorded in vacuum using an alpha spectrometer equipped with a semiconductor detector of 50 mm^2 in area and nominal energy resolution, FWHM, of 10.6 keV. All the equipment is commercially available. The number of the channels used in the measurements was 4096 (energy region 0–6 MeV) and one channel corresponded to 1.5 keV. Neither gain shift stabilization methods nor magnets reducing the coincidence effects were used in the measurements.

3. ADAM program

ADAM treats the alpha peaks of a nuclide as a family. The peak shape model in ADAM is a Gaussian distribution convoluted with up to three low-energy side exponential functions characterized by the slope parameters $1/\lambda$, $1/\mu$, $1/v$ (in keV) and their area ratios R_1 and R_2. The values of the shape parameters depend on the detector in question and on the measurement setup. The peak family areas, the energy calibration, as well as the shape parameters can be fitted individually or together or the user can fix them. The peak shape parameters may also vary depending on the nuclide in question.

All nuclides are fitted simultaneously (Fig. 1). The energy and yield of individual peaks are obtained from the nuclide library data. The complete covariance matrix is calculated during the fitting, which improves the statistical control and produces realistic estimates for the uncertainties. The quality of the fit is indicated with residual and reduced chi-square values.

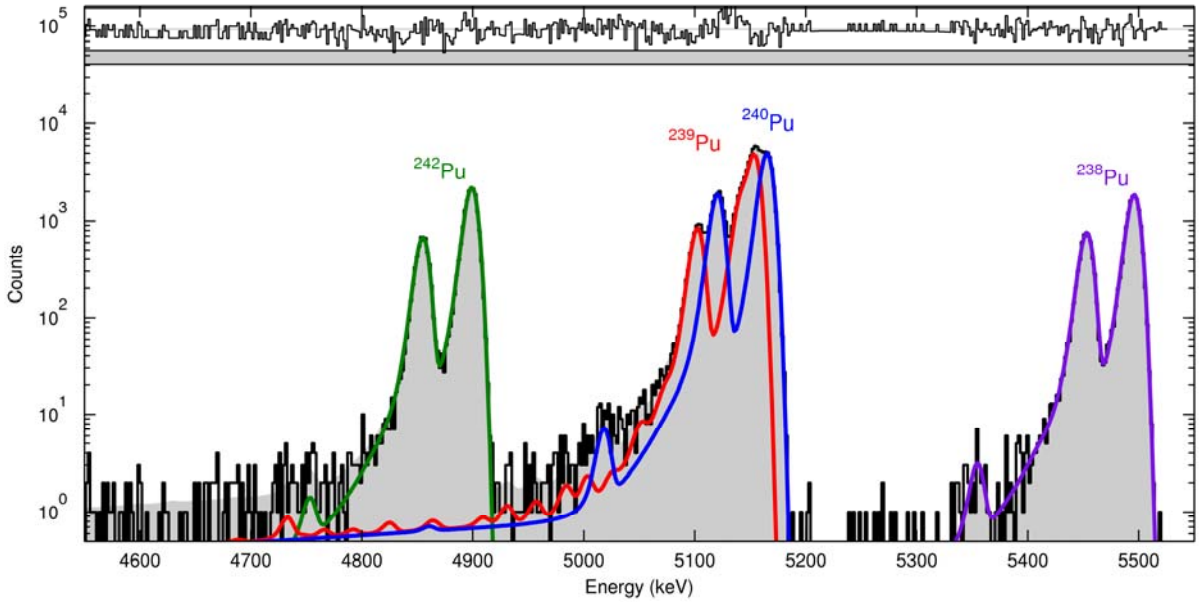

Figure 1: Unfolded alpha-particle energy spectrum from a source containing equal amounts (in terms of activity) of ^{239}Pu and ^{240}Pu. The source also contained ^{242}Pu and ^{238}Pu. Nuclide-specific alpha peak families are presented. The residual is at the top. The measurement was done at source-detector distance of 48.5 mm. The number of counts in the peak family of ^{239,240}Pu is approximately 10^5.

4. Generation of the simulated alpha spectra

A large set of simulated spectra with different statistics was produced to test the precision of the fitting method. The spectra consisted of ^{239}Pu and ^{240}Pu peaks with yields > 0.001. The peaks were generated by using the peak shape parameters listed in Table 1 and dispersing the counts on each channel according to the Poisson statistics.

Shape parameter	Value
FWHM (Gaussian peak)	8.98 keV
$1/\lambda$	3.84 keV
$1/\mu$	16.4 keV
$1/v$	540 keV
R_1	12.2
R_2	0.097

Table 1: Values of the peak shape parameters for a 50 mm^2 detector [3].

5. Analysis of the measured alpha spectra

Details of the analysis procedure were reported in reference [3] and are only briefly summarized here. The data acquisition time (29–197 h) was selected so that the maximum number of counts per channel for the most intense peaks was > 10^3. Three exponential functions were used to account for the peak tailing when the number of counts in the peak family > 10^4. One or two exponentials were used for smaller number of counts. The shape of the peaks of every nuclide was assumed to be equal. Table 1 refers to the average values of the shape parameters.

Second order polynomial was used for the energy calibration. Fitting region of the spectra was 4–5.6 MeV. Background spectra were registered several times. The background was found to be negligible. Nuclide composition, i.e. the information that there are four isotopes of Pu (^{238}Pu, ^{239}Pu, ^{240}Pu and ^{242}Pu) present in the samples, was the only external information used in the analysis. All other information was taken from the spectrum under investigation.

6. Results

The ^{239}Pu/^{240}Pu isotopic ratios obtained by alpha spectrometry were compared to the NIST reference values. The agreement was very good in all six samples as evidenced in Fig. 2.

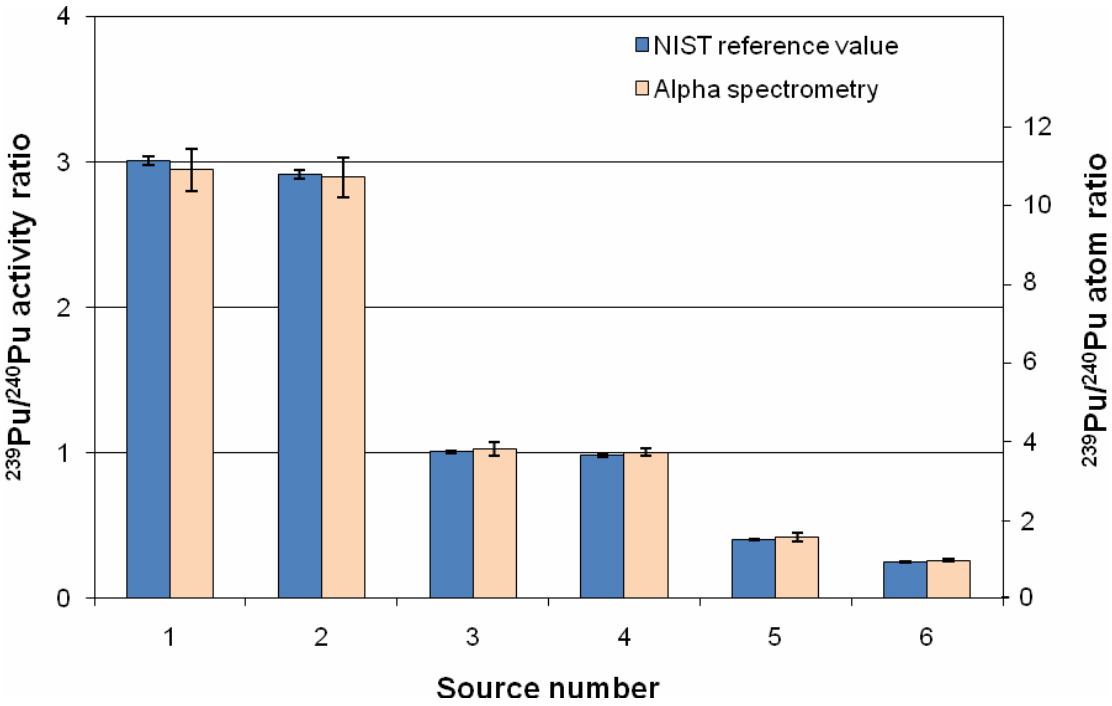

Figure 2: ^{239}Pu/^{240}Pu isotopic ratios of the 6 sources obtained by alpha spectrometry compared to the NIST reference values. The error bars refer to 2 sigma uncertainty.

The uncertainty of the determined ^{239}Pu/^{240}Pu isotopic ratio depends on the counting statistics as well as on the capability to distinguish counts of the nuclides from each other. The contribution of the statistics can be calculated assuming that the number of counts in ^{239}Pu and ^{240}Pu peak families follows Poisson statistics, and thus, $\sigma=\mu^{1/2}$.

The uncertainty of the ^{239}Pu/^{240}Pu isotopic ratio was investigated in the case where the activity ratio is 1 (corresponding ^{239}Pu/^{240}Pu atom ratio of 3.67 or ^{239}Pu/(^{239}Pu+^{240}Pu) atom ratio of 0.78). The solid line in Fig. 3 presents the calculated uncertainty when only counting statistics is taken into account. As can be seen, the statistical uncertainty of the ^{239}Pu/^{240}Pu activity ratio is 1% (10%) or smaller if the number of counts in the ^{239,240}Pu peak family is larger than approximately 10^5 (10^3). Each data point in

Fig. 3 was obtained by analysing 10 000 simulated spectra. The data points present the median of the uncertainties given by ADAM. Because ADAM takes the uncertainties caused by the overlapping peaks correctly into account, the uncertainties reported by ADAM are always larger than the uncertainties calculated from the counting statistics.

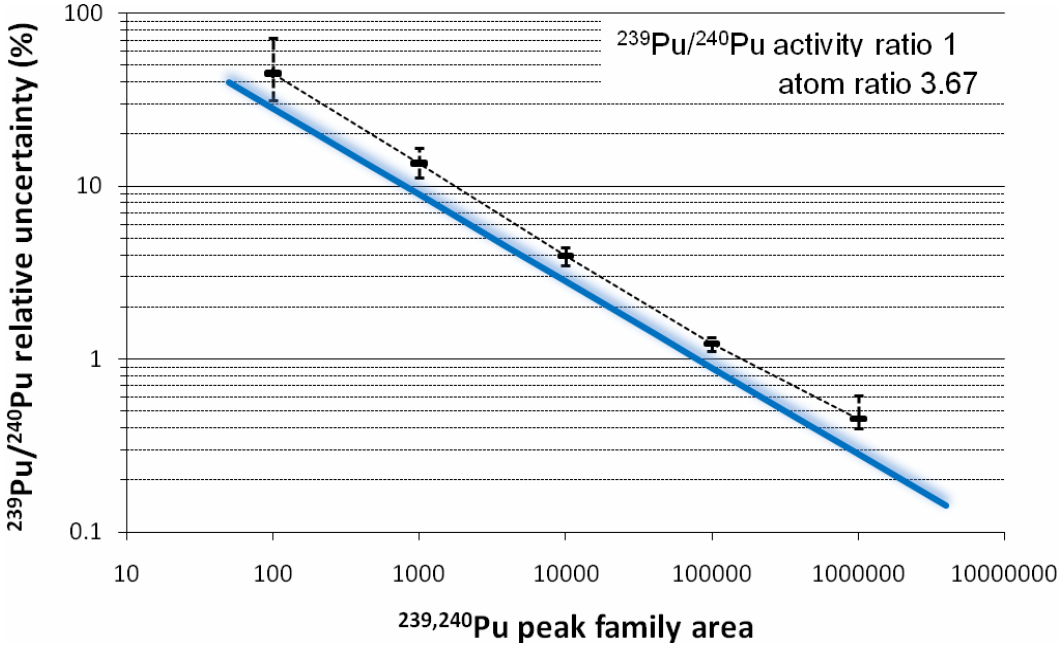

Figure 3: The relative statistical 1 sigma uncertainty of the $^{239}Pu/^{240}Pu$ isotopic ratio as a function of $^{239,240}Pu$ peak family area (solid line). The markers refer to the median of the uncertainties reported by ADAM (dashed line is only a guide for the eyes). Lower and upper limits refer to 5% and 95% fractiles, respectively.

7. Conclusions

High-resolution alpha spectrometry can be used to reliably determine the $^{239}Pu/^{240}Pu$ isotopic ratio. ADAM program for spectrum unfolding produces correct values for peak areas and uncertainty limits.

8. Acknowledgements

K. Inn, J. la Rosa and B.J. Bene from NIST are kindly acknowledged for preparing high-quality sources to alpha spectrometry.

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Development of portable X-ray fluorescence spectrometer for safeguard and forensic analysis

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Abstract

Energy dispersive X-ray fluorescence (XRF) spectrometry is a well known analytical technique for quick, non-destructive, multi-elemental and cost-effective determination of elemental composition of materials. The XRF analysis can be carried out in non-vacuum environment directly on any kind of solid sample without chemical preparation. Safeguard and forensic objects and their measuring procedures are often unique due to necessity of the non-destructive analysis.

The aim of this work is to design and construct a new portable X-ray fluorescence spectrometer which enables performing spot analysis of non transportable or valuable objects on site or in laboratory without any restriction for size, shape and composition.

The new portable XRF spectrometer includes a low-power (4 W) transmission type X-ray tube with Ag anode produced by AMPTEK and a thermoelectrically cooled silicon drift detector (SDD) with 15 mm^2 active area (X-PIPS, Canberra) for detection of characteristic X-rays emitted by the sample elements.

For determination of concentration of sample elements a new calculation model based on fundamental parameter method (FPM) was developed and a new computer code was constructed in MATLAB environment. The fundamental parameter method is typically used for elemental analysis of objects when no standard samples are available.

Measurement and determination of quantitative composition of standard alloys have also been carried out in order to test the reliability of this new FPM based quantitative analysis. Examples of measurement and calculation of concentrations will be presented demonstrating the power and effectiveness of this new spectrometer.

Feasibility-Study on Laser-Induced Breakdown Spectroscopy for Pre-Screening of Environmental Samples

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Abstract:

Among the recent technical developments in Safeguards applications, Laser-Induced Breakdown Spectroscopy (LIBS) provides a new approach for characterization, analysis and detection of nuclear material. In this technique sample material is ablated by an intense laser pulse and information on material composition is obtained by spectroscopic analysis of light, which is emitted from the plasma created by the laser pulse. The LIBS method has the potential to measure isotope ratios of U-235 and U-238, hereby determining the uranium enrichment of the studied sample. In this context the IAEA has identified the need for a pre-screening method for swipe samples obtained by environmental sampling before performing a detailed analysis in the Network of Analytical Laboratories for the IAEA.

As part of the German Support Programme to the IAEA, the Fraunhofer-Institute for Laser Technology is exploring scanning microanalysis LIBS on swipe samples to supplement present analytical procedures. The advantage of the LIBS methodology is that it does not require any sophisticated sample preparation and promises a quick evaluation of uranium isotope ratios of collected material. This paper presents some details of the apparatus and the measurement procedure and discusses first performance results on various types of test-samples containing diluted amounts of uranium. The test samples serve as representation for swipe samples and are prepared with various uranium concentrations in order to assess the sensitivity and dynamic range of the technique. A further topic is the current strategy to address issues like contamination during the analysis.

Keywords: environmental sampling; pre-screening; uranium isotopic analysis; LIBS

1. Introduction

As a strengthening measure of international safeguards, the IAEA acquires swipe samples during inspections as a way of detecting the presence of undeclared material and activities [1]. Swipe sampling is based on collection of dust particles and subsequent analysis of the isotopic composition of collected uranium and/or plutonium traces. A drawback is the requirement of rather sophisticated procedures and instrumentation as this method requires highly sensitive trace analysis in the laboratory. Presently two categories of swipe sample analysis exist: On the one hand bulk analysis of the entire dissolved swipe sample and on the other hand recovery of individual particles from the swipe which are then individually analysed. The main focus lies on the determination of the isotopic composition of uranium, mainly the enrichment of uranium-235. However, minor isotopes like uranium-234 and uranium-236 also serve to indicate presence of undeclared activities. Before the actual trace

analysis is undertaken, swipe samples are pre-screened as a measure to increase the efficiency of the entire work-flow [2].

In 2008, the IAEA organized an Experts and Users Advisory Meeting on Laser-Induced Breakdown Spectroscopy (LIBS) as an effort to identify applications for emerging laser techniques in nuclear safeguards. With support of the German Member State Support Programme the Fraunhofer Institute for Laser Technology located in Aachen, Germany, is undertaking a feasibility study of LIBS as an analytical method for pre-screening of environmental swipe samples at the Safeguards Analytical Laboratories (SAL). The LIBS method allows to perform element analysis (e.g. U, Cm, Pu) of sample material and to determine the U-235 enrichment of collected material without requiring any sort of sample preparation. The ILT study is aimed at determining the sensitivity and dynamic range of the LIBS pre-screening method compared to existing pre-screening procedures currently used at SAL. The application differs from those presented in recent developments of LIBS in nuclear safeguards which mainly focus on portable devices for the identification of nuclear material during inspections [3, 4]. The present paper discusses specific aspects of the LIBS instrumentation for the microanalysis of particles of nuclear materials.

2. LIBS Microanalysis Scanning System

2.1. LIBS Analysis Technique

With the recent advent of robust and reliable pulsed laser systems, the LIBS method is increasingly being established as an analytical device. With this rather recently developed technology the elemental constituents of a sample can be determined, where the sample can exist in solid, liquid or gaseous form. In the case of a solid sample, a short laser pulse of visible or invisible light is focussed onto the sample surface of interest. High irradiation intensities cause the immediate evaporation of surface material. The ablated material creates a vapour above the surface and is additionally heated and excited by the remainder of the laser pulse thus forming a plasma from the evaporated material. In the plasma the compounds released from the sample disintegrate into the atomic elements of which a part is present in its ionized form. These species emit line radiation which is optically guided to a spectrometer for spectral analysis of the individual line components. This way the chemical constituents of the ablated surface can be identified by the element-specific characteristic lines.
Generally, the method is often considered to be non-destructive, or at least minimal destructive, as only small amounts of material are released during ablation. One of the great advantages compared to other methods determining the elemental composition is the fact, that LIBS analysis does not require sophisticated sample preparation and that the analysis provides an immediate result. Therefore the method is well suited for in-line analysis for process control in industrial applications or for the scanning analysis subject of the present paper. At this point it should be noted, that a spectrum is acquired for each laser shot, making LIBS a very “efficient” analysis method. Apart from the ability of identifying chemical elements, high resolution spectrometer systems are able to resolve optical emission from different isotopes. Isotope identification is possible due to the so-called isotopic shift caused by the influence of the nucleus on the atomic energy levels and thereby on the optical light emission properties. By their nature isotopic shifts of the optical emission line are very small (approx. 10^{-5} relative shift).

2.2. LIBS scanning microanalysis

The concept of scanning microanalysis using LIBS has been realised by ILT for production control in industrial applications, e.g., the identification of inclusions in steel samples. Such systems perform up to 1000 measurements on a grid pattern, each one at a different location. Areas of several cm² are mapped within about 10 minutes depending on the step size.
The analysis of particulate matter on substrates using the scanning LIBS methodology has been developed at the ILT for previous studies of airborne particles collected on substrates. The setup used for the studies outlined within this paper was developed for such purposes and has been presented in an earlier publication [5] and is shown in Figure 1. For the safeguards application, swipes samples acquired during the inspection are mounted onto a holder, and short and intense laser pulses probe the surface of the swipe. The sample holder is mounted onto a xy-translation stage, such that the probing laser beam scans along the surface of the swipe sample. Currently an automated raster scan of the surface is envisioned using 100 x 100 steps of 100 µm which results in a total scan area of

1 cm². The all solid-state laser system provides 7 ns pulses in the infrared with a pulse-energy adjustable up to 3 mJ at a repetition frequency of 1 kHz. For the analysis of dust deposited on a substrate it is assumed that single pulses will be applied to each individual position. Whereas fast spectrometers dedicated to the application are used in full-developed scanning LIBS systems, a general purpose spectrometer is used for the current study which is determining the data acquisition time. Since the diameter of the irradiated area amounts to a few tens of µm, approximately 10 per cent of the area is actually incorporated into the measurement. This ratio depends on the step size which can be freely adjusted down to one micrometer. The laser measurement is undertaken in a vacuum-tight analysis chamber such that the sample is studied in a controlled gas atmosphere. Adjusting gas type and pressure gives the possibility to optimize the analysis parameters. Several atmospheres of air, argon and nitrogen with pressures ranging between 1 and 1000 mbar can be adjusted.

The parameters defining the performance of the analysis are on one hand given by the laser system and the interaction with the sample material which determines the amount of light emitted by the plasma generated. On the other hand the spectroscopic unit determines the achievable analytical quality of the measurements. For the specific application trade-offs need to be considered between sensitivity and selectivity which are given by detection efficiency, signal-to-noise ratio and spectral resolution. Additionally, the spectral range covered by the system, timing control of the measurement and data processing capabilities have to be taken into account. For the measurements presented here, a general purpose broadband Echelle spectrometer (LLA GmbH, Germany) equipped with an iCCD detector is employed. It covers a spectral range from 200 to 870 nm with a spectral resolution ranging from 5 pm in the UV to 20 pm in the NIR spectral range.

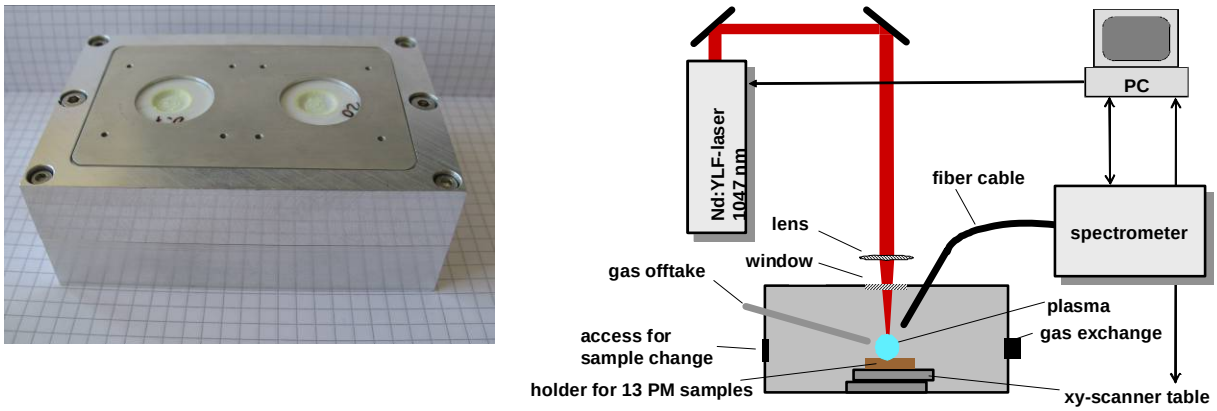

Fig. 1. Left: Sample holder for two ceramic samples with an aperture for each sample of 22 mm diameter, shown without protective windows. Right: Set-up for scanning LIBS analysis of swipe samples.

2.3. Goals/Challenges

Currently, the feasibility aims at determining the enrichment of uranium of approximately 1 µg/cm^2 concentration on the sample surface. A major concern is the presence of spectral lines from other elements which may overlap with the uranium signal. Background mainly originates from the swipe material and the collected dust, where the dominant contribution is seen for Pb, which is strongly present on swipes as it is a common shielding material. A typical dust load is assumed to be 1 mg/cm^2. Another challenge is the demand to resolve the isotopic lines of U-238 and U-235 in order to be able to determine the enrichment of the sample. Therefore sufficient line intensity must be reached, without compromising the resolution through broadening of the line widths. Adjustable parameters are the laser intensity, the ambient gas atmosphere and the iCCD setting of the spectrometer. The feasibility project follows a phased approach based on the following steps:

1. Determination of optimal parameters for scanning microanalysis using prepared samples with an uranium layer of varying enrichment.
2. Measurements on swipe samples: blank and swipes prepared with uranium of varying enrichment and dust
3. Control samples: real swipe samples of known uranium content and enrichment

The first aim is to develop an analysis scheme in order to classify the uranium enrichment into four categories: (I) natural (0.72 %), (II) low enriched uranium (3 %), (III) highly enriched uranium (20 %), and (IV) weapons grade uranium (90 %). A further step envisages the determination of the uranium enrichment from the acquired line spectrum. As line widths and background contribution are yet to be explored, analytical schemes for the enrichment determination as well as the evaluation of achievable accuracy and precision will be subject of work to be conducted during this project. Another question to be answered is the detection limit for uranium, which will be established in the future line of work.

Although LIBS is sometimes considered to be a non-destructive method, the laser pulses leave clear markings on the swipe and the material within the laser focus is released from the surface into the uncovered hemisphere. Because of unavoidable particle release, contamination of the sample chamber needs to be considered and part of the swipe needs to be sacrificed for the analysis. The threat of contaminating the LIBS sample chamber is addressed by collection of released material with a gas and dust off-take via a steady gas flow. The gas flow is guided through a fine dust filter, where the particulate and re-condensed material is deposited and would be available for subsequent analyses. An additional measure includes an exchangeable protective window placed above the sample surface which is transparent to laser-light and emission from the induced plasma. The protective glass window avoids contamination of the sample chamber and can be replaced for each sample.

3. Studies on Test-Samples

The major topic of work at this point is the ability to distinguish the uranium isotopic lines from the optical line emission. As line intensities and line widths recorded in LIBS depend on several parameters like laser-intensity, atmosphere in the analysis chamber, etc. the optimal process parameters needed to be developed which includes extensive measurement series. For these studies, samples of enriched uranium were prepared from nuclear dispersion fuel with two different enrichments in uranium-235, embedded in an aluminium matrix. The advantage of the prepared samples is the stability under laser irradiation as laser ablation induces shock waves in the sample matrix which the fuel dispersion can better withstand than samples with analyte material deposited on the surface. These materials are used to map the most relevant parameters and proof of principle experiments. Presently, two materials with a declared enrichment of 16.1 % and 90.1 % were studied.
The two samples with dimensions of a few millimetres are fixed between two aluminium plates in the samples holder to allow access for the laser measurement to one side of the sample where the uranium compound is accessible at the surface. Figure 2 (left) shows a picture of the metallic U_3Si_2-sample between the back anodized aluminium. Since the material is curved a gap appears between the sample and the holder. The actual sample consists of a surface layer (300 µm thickness) of uranium dispersed in a matrix. The uranium containing layer is supported by a sheet of aluminium (thickness approx. 1.5 mm). A single line scan using LIBS was carried out along the cross section of the sample and the holding plates with a step size of 50 µm. At each point a spectrum was recorded generated by a single laser pulse of 1.5 mJ pulse energy. The spectra are evaluated individually for a number of emission lines, which can be identified using existing data bases like [6]. An example for three emission lines originating from aluminium, silicon and uranium for a line scan is also shown in Figure 2 (right) together with the identified regions on the sample arrangement. The Al emission is present from both the holders and the sample of 1.5 mm width, being stronger on the clean metal sample than on the black anodized holder material. Uranium appears together with silicon only over an approximately 300 µm wide range of the sample, where it is embedded in the aluminium structure. A weak Si signal is also observed from the holder where it is present as an alloying element.

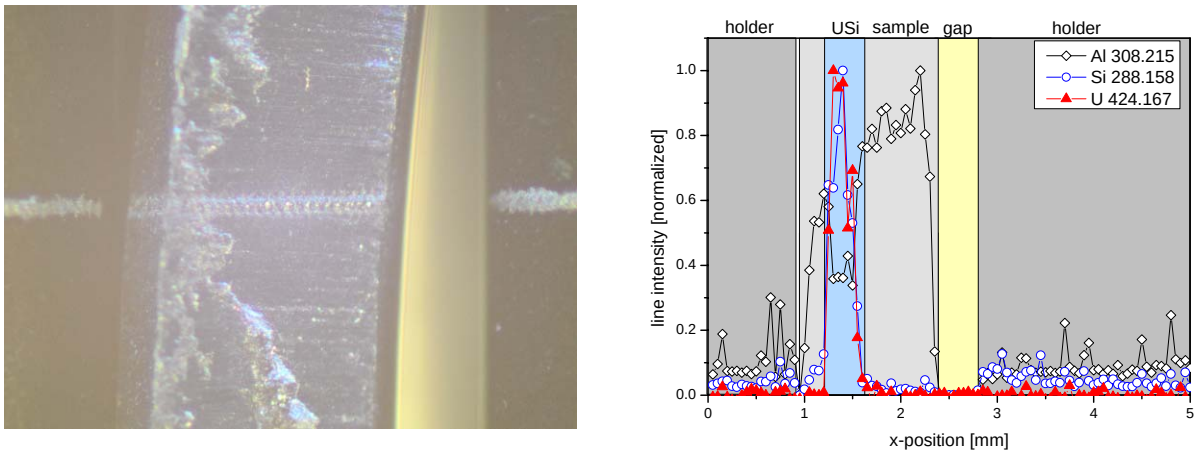

Fig. 2. LIBS scan of the U_3Si_2-sample. Left: microscopic picture of scan trace across the sample. Right: LIBS signal intensities for Al, Si and U as a function of measurement position.

A series of scanning LIBS measurements has been carried with both dispersion samples, which represent different degrees of uranium enrichment. Average spectra are calculated for both materials and excerpts are shown in Figure 3 (left), which can be regarded in a first approximation as representative mainly for one the two major isotopes. Different uranium lines are identified in the spectra and two neighbouring lines from single-ionized uranium, here denoted as U II lines *A* and *B*, are presented in the graph. The lines are identified according to the NIST data base [6] with the wavelengths 424.166 nm (*A*) and 424.436 nm (*B*). The normalized signal curves of line *A* are closely overlapping and have a higher amplitude than *B*. Line *B*, in contrast, shows a significant isotopic shift which clearly exceeds the pixel dispersion of the spectrometer of about 11 pm for this wavelengths range.

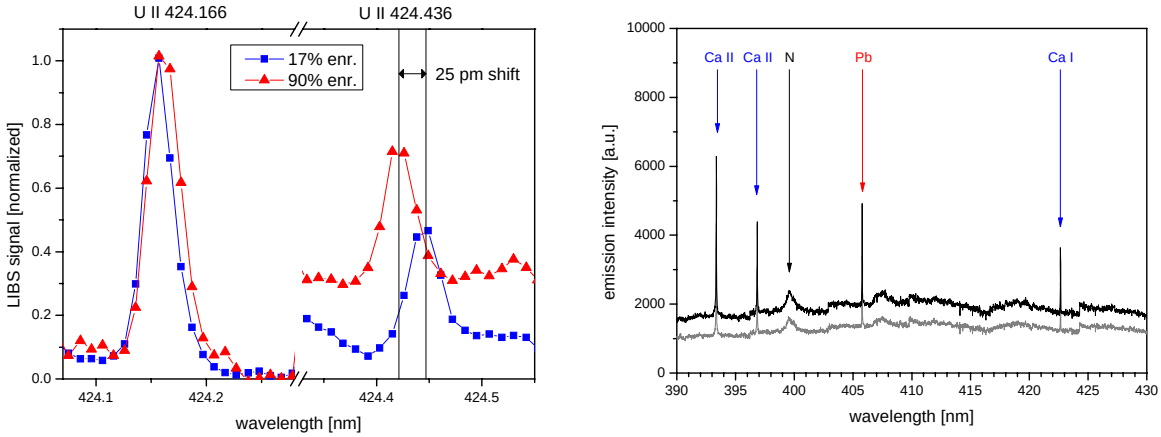

Fig. 3. LIBS spectra of test samples. Left: spectra of U samples with a line shift indicated for the U II line *B* at 424.436 nm. Right: spectra of a Pb swipe sample, including line emissions of Pb at 405.781 nm and Ca I at 422.673 nm. Wavelengths taken from [6].

Spectroscopic details of uranium lines are compiled in [7] and some data for the shown lines are summed up in Table 2. Line *B* is a ground state line with a strong isotopic of the upper energy level, resulting in an isotopic line shift $\Delta\lambda$ (U238-U235) of -25 pm. For line *A* both energy levels are subject to isotopic influences and the resulting line shift is only about 6 pm towards longer wavelengths for U-235. In fact, even the weaker line shift of line *A* with opposite direction when compared to line *B* is slightly indicated in Figure 3.

Although a large number of spectral lines of uranium are known and can be identified in LIBS spectra, these two lines are used most often to study uranium since line *A* is a rather sensitive line suited for trace analysis and line *B* exhibits a relatively strong shift $\Delta\lambda$ useful for isotopic studies [3, 8]. As both lines are closely neighbouring they can easily be observed together when CCD spectrometers are

used. For the SAL pre-screening application the combined usage of both lines offer the possibility to obtain both sensitive and selective measurement results.

Table 2. Spectroscopic line and corresponding energy level data for uranium lines identified in Fig. 3. Data taken from [7].

species, line	λ [nm]	ν_1^{-1} [cm^{-1}]	ν_2^{-1} [cm^{-1}]	E_1 [eV]	E_2 [eV]	$\delta\nu_1^{-1}$ [cm^{-1}]	$\delta\nu_2^{-1}$ [cm^{-1}]	$\Delta\lambda$ [pm]
U II, *A*	424.1664	4585.434	28154.450	0.569	3.491	-1286	-955	5.96
U II, *B*	424.4372	0	23553.977	0.000	2.921	0	-1389	-25.03

To quantify the analytical performance of the measurement technique samples of different levels of uranium enrichment have been prepared as thin layers on a ceramic substrate, also shown in Figure 1. This was achieved by bringing uranium nitrate solution prepared from certified reference material onto the ceramic substrate which was subsequently dried in an additional evaporation step. In an initial classification trial, two samples with natural and highly enriched deposited material have been probed by a series of ten individual LIBS measurements each. The data evaluation provides an overall identification correctness of 90 % for this single pulse microanalysis.

Additionally, test measurements have been performed on swipe samples of lead bricks for an assessment of interfering spectral lines from background material collected with the swipe. Spectra acquired with the LIBS microanalysis system at two selected positions, identified as lead containing, are also shown in Figure 3 (right). They show a few lines of Ca and Pb, which however do not interfere with the uranium lines of interest at 424 nm. This gives a first indication, that critical line interferences from compounds present in dust or the fabric do not have to be expected.

4. Further Steps

The studies of the test samples have demonstrated that the chosen approach has the potential to identify small amounts of uranium and distinguish different levels of enrichment. In the on-going work, the optimal settings for laser intensity and the sample chamber atmosphere will be determined from the series of measurements conducted with the nuclear dispersion fuel samples. Based on these results, a first analytical approach for determination of the enrichment will be developed based on the ceramic substrate samples with the various enrichment levels. The sensitivity and selectivity of the method will be optimized, e.g. by choosing a spectrometer with customized properties especially suited for this particular measurement task.

After showing that LIBS is capable of detecting uranium in the specified concentrations and to differentiate different enrichment levels, real swipe samples will undergo testing, starting with blank swipes, swipes prepared with dust and swipe samples containing uranium particles. The final goal is to analyse realistic swipe samples containing uranium particles and regular dust as background to establish the feasibility of LIBS to be used as a pre-screening method at SAL.

A further line of future work is the evaluation of available measures and equipment to enhance the applicability of the scanning microanalysis LIBS concept for the analysis of uranium residues. When the method will have been established, a design study is planned in order to design a LIBS micro-scanning device which is capable of routinely analysing swipe samples at SAL.

5. Summary and Conclusions

This paper outlines a feasibility study on performing LIBS analysis on swipe samples. A laboratory experiment based on a versatile design for scanning surfaces and conducting elemental analysis via LIBS has been presented. The measurement results obtained on test samples containing uranium of varying enrichment have demonstrated, that the use of LIBS as a scanning microanalysis tool has the potential to analyse microscopic amounts of material for their uranium content and level of enrichment. A first evaluation experiment indicates that there are no severe disturbing interferences from line emission from typical background contributions typically found on swipe samples. Since the analysis of each spot is performed in a few tens of microseconds, the concept is suitable to provide a fast pre-screening system for the analysis of swipe samples for safeguards applications.

6. Acknowledgements

This work is funded under Task A1835 through the German support programme to the IAEA.

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10 Nuclear security and Border Monitoring II

Searching Radioactive Material with Hand-Held Gamma Detectors

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Abstract:

Reliable measurement techniques are decisive for early detection of illicit use of nuclear and radioactive material. In the context of the global threat of nuclear terror, detection of illicit nuclear and radioactive material is of high importance in order to prevent possible terrorist acts with dirty bombs or improvised nuclear devices (IND). Acquisition of nuclear or radioactive material is generally considered to be the major obstacle for terrorists which is most difficult to overcome on their way to assemble and ultimately use a dirty bomb or an IND. Therefore, sophisticated detection methods need to be employed to localize and identify illicit radioactive and nuclear material. As far as border crossings are concerned, portal monitors with large-volume detectors are best suitable for these purposes. But detection of such covert radioactive or nuclear material in places where the material is temporarily stored prior to the composition of the device or detection of the composed device requires a different type of detection equipment. New electrically cooled hand-held systems featuring high resolution gamma spectroscopy with a reasonably low weight to accomplish thorough surveys of suspicious locations became available recently. Germanium detectors with liquid nitrogen cooling were commonly used in that respect before. In case liquid nitrogen is not available, electrically cooled Germanium detectors are the only type of detector to fulfil the requirement of high energy resolution whereas light-weight detectors for on-site surveys are usually equipped with low-resolution crystals. We have performed measurements to investigate the characteristics of such a detector as well as measurements examining the quality of identification by automatic routines of the material in question. The results of identification and search tests will be shown. A comparison with other detectors for mobile search will be presented including an assessment of their applicability for realistic search tasks. New easy to use high-resolution search detectors will be a valuable completion to the widely used low resolution search devices and may thus help to prevent nuclear terrorism at an early stage.

Keywords: illicit trafficking; hand-held device; in-situ measurement; nuclear terrorism; dirty bomb

1. Introduction

Prevention of terrorist acts with nuclear or radioactive material is one of the most important challenges in the field of security today. Sophisticated detection methods are required to localize and identify illicit radioactive and nuclear material. Therefore, either the nuclear material prior to the assembly of the device or the assembled device has to be located at an interim storage place or during transport.

Hand-held detectors play an important role to search relevant areas within a reasonable period of time. Additionally, sometime an identification of the radioactive material in-situ after it's localization is helpful. Most hand-held detectors are equipped with an identification routine, but because of the variation in type and size of detector material as well as the quality of data analysis routines, not all of those detectors are really well suited for identifying nuclear or radiological material. We investigated the usability of several hand-held detectors for in-situ localization and identification of radioactive material.

2. Experiments

We performed search and identification measurements in one of our laboratories to investigate the performance of hand-held detection systems in an on-site survey [1]. Because of the small dimensions of the laboratory (L x W X H = 8 m x 4 m x 2.5 m) the search could only be performed on a relatively small area. Figure 1 shows a picture of the laboratory, taken from the entrance door.

Figure 1: Laboratory where the radioactive source was hidden.

A Co-60 source with an activity of 350 kBq was used to represent the hidden radioactive material. In this type of source the radioactive material is embedded in a transparent chip of Lucite. Because of its small dimensions (diameter: 2.5 cm, see figure 2) and transparence it was relatively inconspicuous and difficult to spot by the eye. It was therefore well suited for this study because the general aim of the investigation was to figure out if and how quickly test persons would be able to locate the source just based on the detectors' readings and signals.

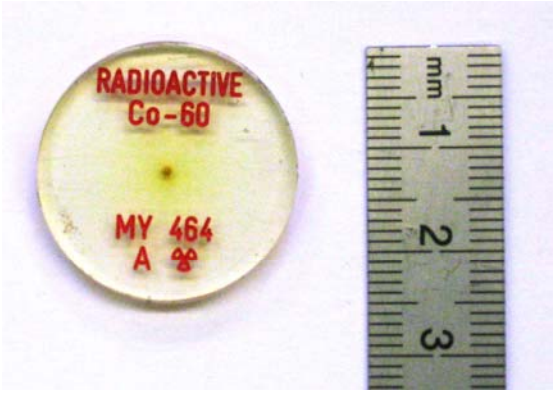

Figure 2: Co-60 source used for the searches.

Five search and identification detection devices by four different manufacturers were investigated, featuring different crystal materials, sizes, weights, and analysis routines. In addition, the dose rate meter FH40 which is widely used was investigated for comparison. Figure 3 shows pictures of these detectors. Their relevant specifications are listed in table 1 (see also references 2-8).

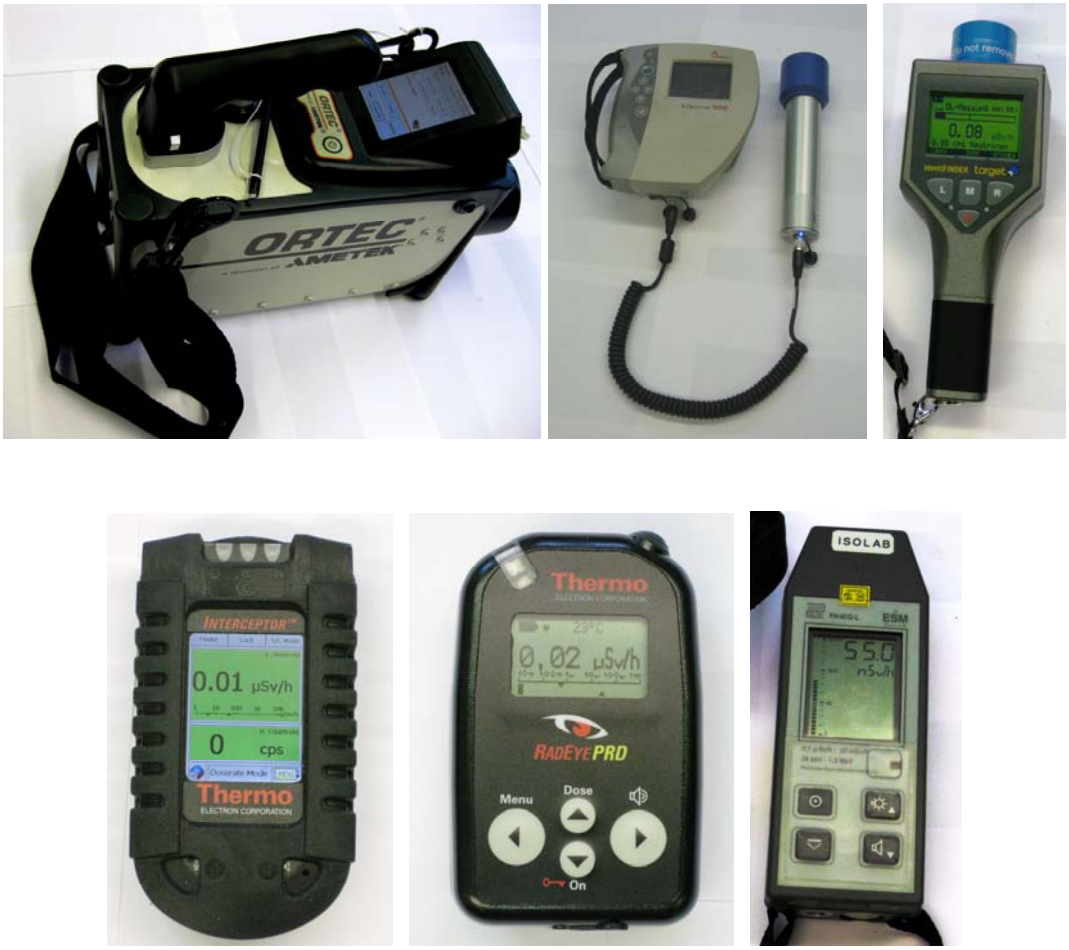

Figure 3: Detectors used, top left: Micro Detective, top center: InSpector 1000, top right: IdentiFINDER, bottom left: Interceptor, bottom center: RadEye PRD, bottom right: FH40 G-L.

In order to gain comparable results for each detector we selected hideouts for the source located at four different heights and had test persons searching the hidden source once for every height with each detector. This way we had 24 search runs for every test person. The chosen heights were:

1. 220 cm – 232 cm
2. 123 cm – 130 cm
3. 77 cm – 98 cm
4. 30 cm – 44 cm

The range of heights within a specific group was due to the possible hiding spots in the laboratory which did not allow us to choose hideouts at the exact same height, but we attempted to keep the ranges as narrow as possible. The order of the hiding spots selected in each series of search runs was altered every time. In this way we assumed to achieve a higher degree of objectivity as far as the comparison of the detectors' performances was concerned.

The surveys always started at the laboratory's entrance. This was also taken as the starting time of the measurement. The way through the room and also the walking speed during the survey were

chosen by the test person. Among those persons were people experienced in handling radiation detectors, but also people who did not know the detectors at all. This way of performing the surveys included several random factors due to the individual preferences of the test persons. Nevertheless, in a real situation different strategies would have to be expected as well, even if only experienced people were involved. Figure 4 shows two of the test persons right after locating the source in two of the places selected prior to the surveys.

Table 1: Comparison of detector specifications.

Detector	**Micro Detective**	**InSpector 1000**	**IdentiFINDER**	**FH40 G-L**[1]	**RadEye PRD**	**Interceptor**
Manufacturer	Ametek / ORTEC	Canberra	ICx Radiation	Thermo	Thermo	Thermo
Detector Material	HPGe	$LaBr_3$	NaI (Tl)	Gas (PC)	NaI (Tl)	CdZnTe
Weight [kg]	6.9	2.4 (Body + Probe)	1.25	0.45	0.16	0.27
Size of Device [cm]	37.4 x 14.6 x 27.9	19 x 16.5 x 6.4 (Body)	24.8 x 9.4 x 7.6	19.5 x 7.3 x 4.2	9.6 x 6.1 x 3.1	11.2 x 6.1 x 2.5
Crystal Size [cm]	3 (Length) / 5 (ø)	3.8 (Length) / 3.8 (ø)	5.1 (Length) / 3.6 (ø)	2.6 (Length) / 2.5 (ø) (Gas Chamber)	3.1 (Length) / 1.8 (ø)	0.7 x 0.7 x 0.35
Identification Mode	yes	yes	yes	no	no[2]	yes
Energy Resolution [keV] at 662 keV	1.5	23.2	48	-	-	19
Relative Efficiency [%]	10.7	12.6	8.0	-	-	0.02
Battery Life [h]	> 3	9	8	> 250	600	10

[1] Dose rate measuring device

[2] The RadEye is equipped with the NBR (Natural Background Rejection) routine which enables the user to distinguish between “high energy” and “low energy” alarms.

The surveys were performed by seven test persons which had a different amount of experience with searches for radioactive material. A larger number of people which of course would have been preferable was not available because of the laboratory’s access restrictions as a radiation controlled area for handling radioactive material. The task of the test persons was to locate the source as quickly as possible. Thereafter, the identification routine of the device should be run if the device was equipped with such a routine. The time necessary for locating the source was noted and a “quality factor” assigned which referred to the fact how close the actual position of the covert source was judged by the test person. For two of the detectors an exact runtime for the identification routine had to be set (IdentiFINDER, InSpector 1000), the Interceptor chooses the runtime automatically, and the Micro Detective’s identification routine keeps running until it is manually stopped, showing results of identified nuclides during the process.

Figure 4: Test persons locating the source hidden in a cable rack (on the left) and in a cardboard box (on the right) at different heights (source position marked with arrows) using the Micro Detective (on the left) and the Interceptor (on the right).

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3. Results

Because of the small number of test persons we had to accept large variations of the measurement results. These variations occurred because of the different characteristics of the devices as well as the test persons' individual behavior such as search strategy, e.g. walking speed, walking directions, etc., and personal capabilities. Figure 7 illustrates the results of the mean search times including all detectors and all search results.

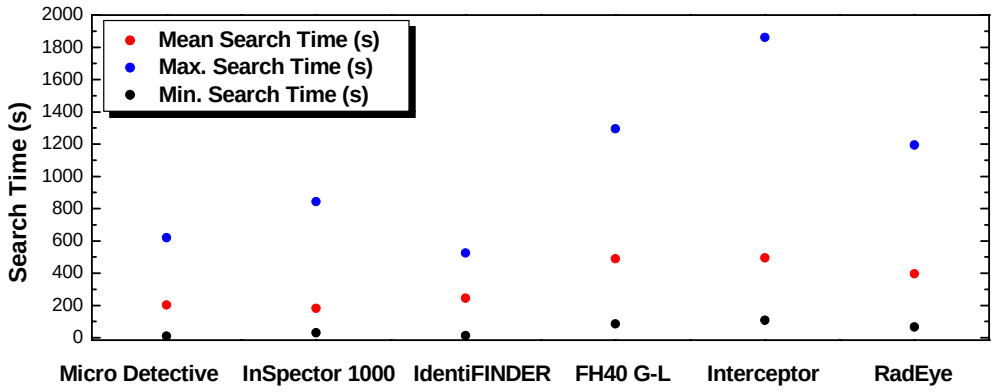

Figure 7: Mean, minimum, and maximum search time for all detectors and all groups of height. In two cases the source was not found at all; these data were not accounted for in the diagram.

Instead of error bars, the maximum and minimum search times for the detectors are shown as well. The search times for the detectors in some cases varied with different heights. As an example, figure 8 shows the mean search times for the surveys where the source was hidden at a height in the range of 77 cm – 98 cm.

Identification measurements were performed with the IdentiFINDER, the InSpector 1000, the Interceptor, and the Micro Detective. As for the former two, the runtimes for these measurements were set to values considered to be typical by the manufacturers. In the case of the IdentiFINDER, we selected 30 seconds of measuring time. For the InSpector 1000 we chose 120 seconds. The RadEye and the FH40 G-L do not provide identification capability as they cannot measure energy spectra.

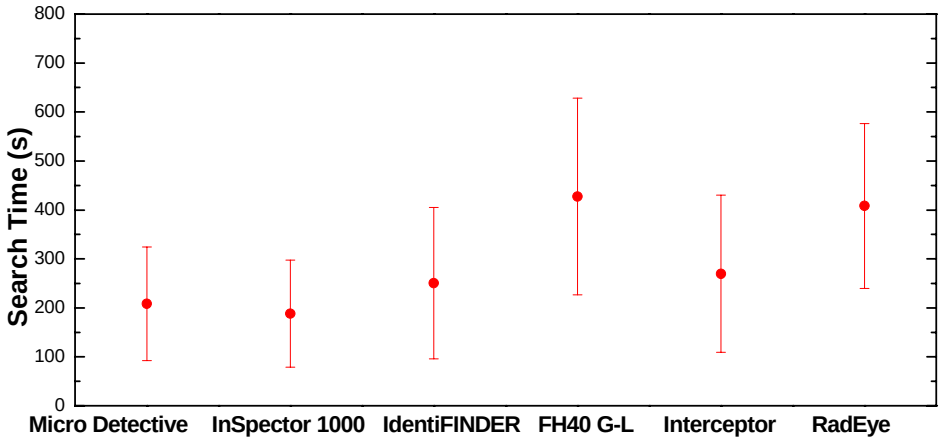

Figure 8: Mean search times for hiding spots at heights in the range of 77 – 98 cm.

The Micro Detective merely discriminates between two “confidence levels” referring to the fact that a certain nuclide is “suspected” or “found”. As for the other three detectors, a “confidence factor” given in percent is shown after the end of the measurement, specifying the certainty of a nuclide being identified. As for the Micro Detective we associated “found” with 99.9 % as the manufacturer suggested.

A comparison of the detector’s runtimes required for the identification of the Co-60 source would not have been informative as some runtimes were set and others were variable (2-144 s). So only the confidence factors given by the detectors were compared. Additionally, we were interested in false identification results given by the detectors because in a real situation users have to rely on the fact that a certain nuclide identified by a detector is really present. Figure 9 illustrates the comparison of the mean confidence factors for all four detectors as well as the number of measurements where false identification results were given.

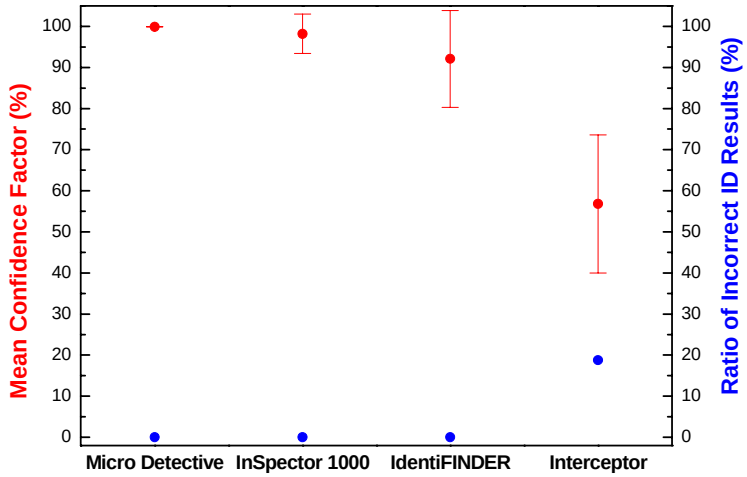

Figure 9: Mean confidence factors for all detectors featuring an identification mode (red dots) and of ratios of false identification results (blue dots).

The mean confidence factor was calculated from all measurements with each detector. Only in the case of the Interceptor false results occurred. This detector also showed the lowest mean confidence factor. Taking into account that the Co-60 source should have been quite simple to identify at near distance the Interceptor's factor of approximately 57 % is very unsatisfactory, but an experienced user can look at the energy spectrum and thus can gain further information. The three other detectors turned out to be sufficiently reliable and did not show any false identification results at all.

In general, the Micro Detective, the InSpector 1000, and the IdentiFINDER are significantly superior to the other three detectors in localizing the source, the InSpector 1000 showing the shortest mean search time of all detectors. This fact refers to the analysis of all surveys and also the surveys of the specific height mentioned above. However, one has to take into account that the detectors are drastically different in weight and therefore in handling. The duration of a survey performed with the RadEye can vastly exceed that of a survey done with the Micro Detective because the latter has approximately 40 times the weight of the former (RadEye). On the other hand, the Micro Detective features the option of identifying localized material with a high degree of certainty which could be immensely valuable in a real scenario. So the best possible choice of detector in case no preliminary knowledge about the material hidden within a certain area would probably be a relatively light-weight detector equipped with an identification mode showing satisfactorily results.

4. Conclusion

We investigated the qualification of five hand-held detectors and one dose rate measuring device with regard to fast detection and reliable identification of hidden radioactive material. As for the localization, all detectors are equipped with acoustic signals which for most of the searchers are more suitable for finding the approximate location of the source than the display readings. To find the exact position, the gamma count rate or dose rate figures shown on the displays are useful as well. The dose rate meter FH40 G-L which was included in the study for comparison with a very common radiation protection instrument proved to be extremely difficult to use for searching. Its response when coming close to the hidden source was very slow, leading to irritations whether suspicious material had been localized or the readings just referred to a small variation in background radiation. Additionally, averaging procedures at low dose rates in the software of the instrument complicate the interpretation of the readings in respect to the direction of the source of radiation. In two cases the source could not be localized at all which did not happen for any other detector. In short, the FH40 G-L dose rate meter is not well suited for this type of surveys.

The search results for the two pocket-sized instruments Interceptor and RadEye were better than those of the FH40 G-L. These two detectors feature acoustic signals that turned out to be more helpful for the localization than the signal of the FH40 G-L. Their low weight allows for long-time search runs. This fact may convince users to choose these detectors despite the limited possibility to identify radioactive or nuclear material (in case of the RadEye) or the flaws of the Interceptor's identification mode whose results were clearly worse than the ones of the Micro Detective, the InSpector 1000, and the IdentiFINDER.

The three latter detectors are equipped with identification modes showing satisfactory results in the case of the Co-60 source which should have been quite simple to identify. The InSpector 1000's option of holding the probe separate of the detector's body proved to be useful, especially for localizing the source in the highest and lowest positions. The holding position recommended by the manufacturer turned out to be tiring in the long run, though. Even its relatively moderate weight of 2.4 kg has a tiring effect in long-time surveys, as well. This also applies to the IdentiFINDER because its mass is not perfectly balanced, and especially for the Micro Detective which is by far the heaviest of the detectors and is therefore difficult to handle at high positions.

In the recent past hand-held radiation detectors have clearly improved. However, a general recommendation concerning the detector that would be the best choice for on-site surveys is difficult to give. There is definitely no "ideal" or "perfect" choice. The qualification and preferences of the search person, e.g. concerning the use of acoustic and/or optical signals, as well as the requirements of the task, e.g. search area dimension and accessibility, are important and appear to have a considerable influence on the detector which is considered to be the "best" one. Some detectors are more suitable for users experienced in radiation measurements. Prior to the surveys, we had each

search person test the functions of the detectors in the presence of the Co-60 source. For example, it is important to be familiar with the characteristic response time of the detector and the location of the detector's sensitive point. This turned out to be very useful for the surveys.

The light-weight detectors Interceptor and RadEye which allow for long-time surveys have disadvantages concerning the average time necessary to localize radioactive material. The Interceptor features an identification mode showing flawed results, the RadEye has no identification mode at all. The Micro Detective, the InSpector 1000, and the IdentiFINDER showed the best results concerning the localization time. Their identification results proved to be correct, too. With these three detectors, however, one cannot search an area as large as that which can be covered when measuring with the Interceptor or RadEye, respectively, because of the differences in weight. A simple dose rate measuring device like the FH40 G-L, in our opinion, is not suitable for on-site surveys. If radioactive or nuclear material is suspected within a certain area and no further information is given about the location, a medium-weight detector featuring a reliable identification mode such as the InSpector 1000 or the IdentiFINDER would probably be the best choice of instrument. A user experienced in radiation measurements, however, may prefer using the Micro Detective despite its heavy weight if the search area is reasonably small.

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ESARDA 2011
Nuclear safeguards implementation, Illicit Trafficking, Oral presentation

Figure of Merit for Evaluating the Performance of Radionuclide Identification in Portal Monitors and Handheld Devices

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Abstract:

The instruments used in the control of illicit trafficking of radioactive materials, either in radiation portal monitors, hand held radiation identifiers, or search systems, are constructed in widely different ways with widely varying detector materials and analysis software. However, within the various groupings (e.g., portal monitors), all instruments are expected to solve the same problem, that is, to detect and identify any radioactive material present according to the prescribed investigation methods (CONOPS). The best way to compare performance of the instruments is with a numerical score or Figure of Merit (FOM). The FOM must quantify the performance of the instrument with respect to false positives (FP) and false negatives (FN). The minimization of FN for certain radionuclides (e.g., SNM) is more important than the minimization of FN for non-threat nuclides (e.g., low amounts of NORM). Likewise, the minimization of FP for SNM is also more important than allowing false reporting of the common NORM nuclides. The performance depends on the details of the testing, so the CONOPS must also be included in the statement of the FOM. We have developed a FOM based on the number of true positives (TP), the number of false important positives (FIP), the number of FP, the number of true positives for SNM (TPSNM), and the number of false positives for SNM (FPSNM). This formula rates the overall performance with extra weight given to FP and FN for SNM. An Example will be shown for testing of both a NaI and HPGE portal monitors in different modes, nuclides, and shielding.

Keywords: FOM, Figure of Merit, HPGe, NaI, illicit trafficking, detection limit

1. Introduction

The need to increase nuclear security and monitor radioactivity in the environment has resulted in the development of many different solutions designed to solve the same problem, that is, detect small amounts of specific nuclides in an essentially uncontrolled measurement situation with high reliability and minimal inconvenience or interruption of progress to the people or objects being monitored. Typically, the time for measurements is short, perhaps less than a minute. Mixtures of nuclides are expected as well as people who have recently had nuclear medicine procedures. For the majority of the users of these monitoring systems, it is not possible to perform testing of the available systems to determine the efficacy of a system in the expected conditions at the monitoring point. To aid the users in the evaluation of systems, others have developed scoring methods for reporting performance [1]. In this work, we describe a scoring system (Figure of Merit or FOM) which is intended to result in a simple numerical score for each system in a prescribed test.

Each test is the data collection, analysis, and reporting that results in either a negative (no activity) or a positive (activity) result. There is one set of results per test which is the list, if any, of nuclides found

during the test. A test can be repeated and the FOM reported on the average score of all the tests. In this work, each test was repeated 32 times.

The result of the test consists of True Positives (TPs), False Positives (FPs), and False Negatives (FNs). A TP with a confidence level above a threshold will create an alarm. Other conditions causing alarms are incorrect time of measurement or speed, excessive gross count rate (high dose rate), or system malfunction. In addition, the term “nuisance alarm” has been used to describe the reporting of any nuclide that is actually present, but whose presence is not of concern. An example is ^{40}K in kitty litter. Another possible report is “irrelevant TP” where the nuclide is present in the background and may be reported by some sensitive devices or long counting times and not reported in other conditions.

A TP is the reporting of a nuclide being detected when the nuclide is actually present in the test situation. A FP is the reporting of a nuclide being detected when the nuclide is not present. A FN is failure to report a nuclide, when the nuclide is actually present in the test situation. However, some TPs and FPs are more important than others. For example, a positive report of plutonium detection is more serious than a report of ^{40}K or other Naturally Occurring Radioactive Material (NORM).

In this FOM formula, the nuclides are divided into categories: Special Nuclear Material (SNM), Important, and Other. Different weighting factors are assigned to each category.

The main purpose of the instrument is to maximize number of TPs. In any monitoring situation, FPs causing an alarm must be minimized because each FP requires an investigation and resolution of the alarm. In this calculation of the FOM, the FPs for some nuclides are given more weight than FPs for other nuclides. Likewise, FNs must also be minimized because this is a failure of the monitoring system to perform its primary function.

The FOM is normalized by dividing by the value of the FOM for a perfect score and scaled by 10. That is, the weighted score for all the possible TPs in the test. This gives the best score as 10, but with no lower limit as FPs and FNs have negative factors.

The intent is that a high FOM will mean that the monitor is performing better in the stated test conditions than a monitor with a lower FOM. Here “better” includes all factors, including interruption to flow of traffic because of the need to resolve FPs as well as the ability to detect the nuclides present in a variety of conditions.

Two different special situations are important: mixtures of nuclides (including spoofing, where a mixture of nuclides is used to mimic the response in the detector of a specific nuclide) and masking of one nuclide by a large amount of another nuclide or nuclides. The performance when multiple nuclides are in the test mixture is accounted for in the scoring as the combination of TPs, FPs, and FNs. The masking situation is different in that the masking nuclide(s) could be NORM, which is often detected in many common materials. To account for

Table I

SNM	Important	Other
^{233}U	^{57}Co	^{18}F
^{235}U	^{60}Co	^{67}Ga
^{237}Np	^{133}Ba	^{51}Cr
Pu (^{239}Pu)	^{137}Cs	^{75}Se
Elevated uranium	^{192}Ir	^{89}Sr
U^{232}/Th^{228}	^{204}Tl	^{99m}Tc
SNM	^{241}Am	^{103}Pd
		^{111}In
		^{123}I
		^{125}I
		^{131}I
		^{153}Sm
		^{201}Tl
		^{133}Xe
		Natural
		^{40}K
		*^{226}Ra
		^{232}Th and daughters
		^{238}U and daughters

masking, nuclides can have multiple alarm thresholds. This will allow small quantities of the nuclide to pass without alarm (not counted as either TP or FN), but should cause an alarm if the level is above the higher threshold (should be TP). If the test condition contains a high level of a “masking nuclide”, the result is a TP if the detected level of the nuclide is above the threshold and a FN if the nuclide is not reported (i.e., no alarm). For these tests, “high NORM” is defined as the emissions from 5 tonnes of KCl stacked uniformly on the entire bottom of a single 8 x 8 x 20 foot CONEX container.

To properly compare the performance of different systems, the details of the test conditions (CONOPS) must be included with the FOM. For example, in freight vehicle portal monitors, the transit speed is important as it determines the length of time for data collection from the container passing through the monitor.

Table I shows the categories, the nuclides in each category, and the weighting factors for the test results shown later. In the situation where a table entry (e.g., Elevated uranium) is reported as a result of the detection of a specific isotope (i.e., ^{235}U) only one result is included in the FOM. In thisexample, however, if elevated uranium was reported with no other uranium isotope reported, it would be counted as a TP or FP according to the test situation.

Table II shows the weighting factors for the different results.

Table II Weighting Factors					
SNM		Important		Other	
TPs	FPs	TPi	FPi	TPo	FPo
4	-4	3	-3	1	-1

The FOM is calculated as shown in the equation below:

$$FOM = 10 * \frac{\sum_{SNM\ found} TPs + \sum_{SNM\ not} FPs + \sum_{Imp} TPi + \sum_{Imp\ not} FPi + \sum_{O} TPo + \sum_{O\ not} TPo}{\sum_{SNM\ Actual} TPs + \sum_{Imp\ actual} TPi + \sum_{O\ actual} TPo}$$

2. Equipment and Setup

The test results shown are based on the ORTEC Detective SNM Portal and the Thermo ARIS-1024 Portal. The ORTEC portal consists of two columns of 4 detectors each. The HPGe detectors are IDMs, as shown in Fig. 1. The Thermo portal consists of two columns each with 8 large volume NaI detectors. The NaI detector module is shown in Fig. 2.

A complete description of the ORTEC portal is given in [2].

One ORTEC monitor is installed (October 2009) at the Anthony, New Mexico Port of Entry and a second one is installed (April 2009) at the ThermoFisher facility in Oakwood Village, Ohio. The New Mexico portal is in operation on commercial truck traffic on Interstate 10 as a secondary portal. The ORTEC portal is in operation as a test facility with controlled sources, shielding, and NORM. The following discussion applies to both units, but the results are only from the test facility. Figure 3 shows the New Mexico installation.

Figure 1 Interchangeable Detector Module (IDM)

Figure 2 NaI detector module

The Thermo ARIS-1024 is installed and operational in many places. The tests were conducted on the ARIS-1024 installed at the Thermo Fisher portal test facility in Oakwood Village, Ohio. Figure 4 shows the ARIS-1024 in a typical setup.

The tests consisted of three different nuclides (^{57}Co, ^{133}Ba, and ^{137}Cs) mounted, one at a time, in a standard 26 ft box body truck, with and without shielding and NORM. The truck was driven at 8 kph through the portal. Tests with these nuclides, various shielding thicknesses, and with and without NORM were performed.

Figure 3 Installed HPGe Portal at Anthony, New Mexico

The truck occupancies for this testing were determined by optical entry and exit sensors positioned about 4 m apart and centered on the detector position. Independent sensors were used for each portal.

The two portals have the same horizontal spacing (about 5 m) and are installed along the same traffic lane. In this way the same test vehicle could driven past both portals at a constant speed in rapid succession to obtain results that were as comparable as possible.

Figure 4. Typical installation of ARIS-1024 portal monitor.

2.1 Source and attenuators

For the example here,^{133}Ba was selected. This source closely resembles Pu-239, an SNM material. The sources used were point sources. The sources were mounted in a wooden frame. The attenuators were 3.2 mm thick steel plates which could be combined to give total thicknesses of 0.32 to 10 cm. The attenuators were placed so as to shield the complete field of view on both sides of the portal from the source. The ^{133}Ba source was 148 µCi, as prescribed in ANSI N42.38.

The NORM was placed evenly on the floor of the cargo section of the truck to give an increased background, but was piled low enough so that it did not block the direct path for gamma rays between the sources and the detectors. The NORM was commercial water softener salt substitute or potassium chloride. About 3 tonne of KCl in 18 kg bags on pallets were in the truck.

3. Results

The ^{133}Ba source was placed in the truck and driven through the portals at about 8 kph 32 times for each thickness of steel shielding. The shielding thicknesses were from 1.27 to 4.5 cm. The nuclides detected were recorded and the FOM calculated for each shielding thickness using the nuclides in Table I and the factors in Table II.

To show the efficacy of the FOM method, the results of only the true positives, that is, only the detection of ^{133}Ba is shown in Fig. 4, where the FOM(TP) is plotted as a function of steel shielding thickness. In this case, only ^{133}Ba is accepted as a TP Important. FPs are not included in this calculation of the FOM.

Figure 5 shows the ARIS-1024 portal does detect ^{133}Ba in the pass through mode for shielding up to 4.1 cm, then the detection drops off. The reduction in the FOM at 3.8 cm is possibly due to a detector failure. The failure was reported by the software.

The HPGe results in Fig. 5 show a steady decline in detection from 100% at a thickness of 2.5 cm to no detection at 4.5 cm. The 3.8 cm data point is also (seeARIS-1024) lower than expected so there may be some unknown factor affecting the detection. The average speed for this point is within the range of average speeds for all the other points.

Figure 6 shows the full FOM, from the equation above, for the same set of tests. Note that the vertical axis ranges from -15 to +15.

In this figure, the ARIS-1024 results show the impact of including the reporting of nuclides not in the truck, that is, FPs. The ARIS-1024 reported FPs for both SNM and Other nuclides.

The HPGe results (full FOM) are similar to the TP only FOM results, indicating a much lower number of FPs. The improvement is largely due to the better energy resolution.

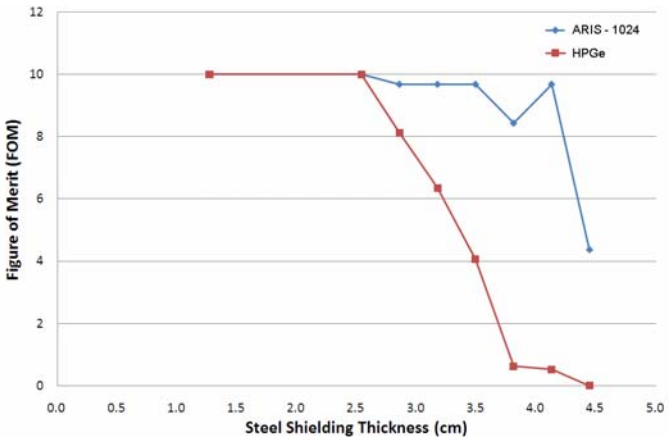

Figure 5. Plot of FOM vs Steel Shielding Thickness for Two Portal Monitors showing results for True Positives only.

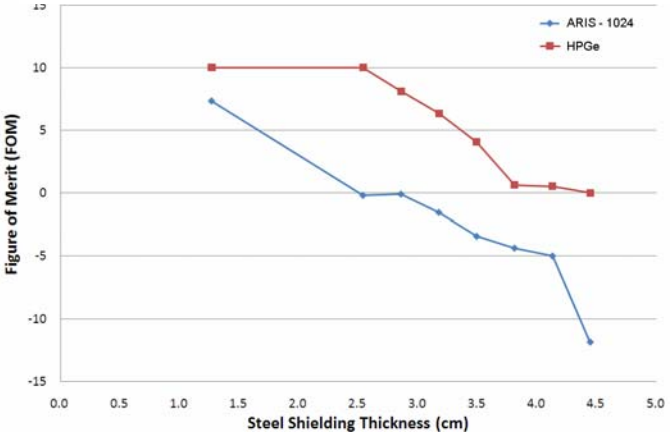

Figure 6. Plot of FOM vs Steel Shielding Thickness for Two Portal Monitors showing results of the FOM as defined.

4. Conclusions

The FOM described in this paper has been shown to provide data that assists in the choice of instrumentation to meet specific security monitoring requirements. This FOM accounts not only for the positive detection of nuclides present, but also the false reporting of nuclides not present, making it useful in the comparison of differing systems designed to solve the same problem. It also provides a useful indication of the extent to which the efficacy of a given system will be impacted by FPs, which must be investigated before the shipment can be released. This FOM applies to any monitoring test comparison, not just portal monitors, because it only uses the results reported by the instrument. In using the FOM, the complete details of the test situation must be given and be as consistent as possible for the different instruments. Future work will include future work with different radionuclides and measurement scenarios. These data will assist the correct choice of instrument for different performance needs.

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Determining the Effect of Concrete Roadways on Gamma-ray Background for Radiation Portal Monitoring Systems

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Abstract:

The dissolution of the Soviet Union coupled with the growing sophistication of international terror organizations has brought about a desire to ensure that a sound infrastructure exists to interdict smuggled nuclear material prior to it leaving its country of origin. To combat the threat of nuclear trafficking, radiation portal monitors (RPMs) are deployed around the world to intercept illicit material while in transit by passively detecting gamma and neutron radiation. Portal monitors in some locations have reported abnormally high background counts with little or no consistency. The higher background data has been attributed, in part,to the concrete surrounding the portal monitors. Higher background increases the minimum detectable activity and can ultimately lead to more material passing through the RPMs undetected.

This work was focused on understanding the influence of the concrete surrounding the monitors on the total gamma-ray background for the system. The study combined destructive and nondestructive analytical techniques with computer simulations to form a qantitative model that is adaptible to any RPM configuration. Six samples were taken from three different composition concrete slabs. The natural radiological background of these samples was determined using a high-purity germanium (HPGe) detector in conjunction with the Canberra In-Situ Object Counting System (ISOCS™) and Genie™ 2000 software packages. The composition of each sample was determined using neutron activation analysis (NAA) techniques. The results from these experiments were incorporated into a Monte Carlo N-Particle (MNCP) photon transport simulation to determine the number of gamma-rays from the different concrete slabs detected by the RPM.

Keywords:smuggling; trafficking; NAA; RPM; concrete

1. Introduction

The collapse of the Soviet Union in 1991 ushered in an era of uncertainty concerning the security of the radiological and nuclear material holdings of the Russian Federation along with the other countries of the Former Soviet Union (FSU). In addition, the 11 September 2001 terrorist attacks on the United States highlighted the growing sophistication of international terrorist organizations, along with their desire to inflict mass civilian casualties. The combination of possibly unsecured radiological and nuclear material, porous borders throughout the FSU, and terrorist organizations seeking to acquire such material, makes radiation detection and ports of entry (POEs) a vital step in combating nuclear smuggling.

From 1991-2006, more than 40% of reported illicit nuclear material trafficking cases had a nexus in Russia and the FSU [1]. To reduce this threat, the United States Government (USG) has engaged in multiple partnerships to provide radiation detection material – including radiation portal monitors (RPMs) – for use at POEs to intercept smuggled radiological or nuclear materials.

RPMs are designed for vehicular, rail, or pedestrian traffic, but in each case they are

used to passively detect gamma and/or neutron radiation. Gammas are detected in the RPMs using any number of different detector technologies; however, this research was specifically focused on gamma detection in vehicular RPMs using polyvinyl toluene (PVT) scintillators. The methods described in this paper may be adapted for any RPM configuration.

When not occupied by a vehicle, RPMs continuously measure the background radiation in the surrounding areas and adjust the alarm thresholds accordingly. In some locations, the RPMs reported abnormally high gamma-ray background with little or no consistency. It is possible that the natural occurring radiological material (NORM) in the concrete roadways beneath the RPMs is contributing to this anomalous data. Higher background levels will increase the minimum detectable activity (MDA) of the RPMs, thereby raising the threshold for radiation alarms and increasing the probability of illicit radiological or nuclear materials passing through the portals undetected. A picture of the RPM modeled for this research is shown in Figure 1.

Figure 1: The RPM modeled for this research.

2. Experimentsand simulations

The primary objective of this research was to develop a method for determining the natural radiological background and elemental composition of the concrete underneath and around an RPM in order to estimate its contribution to the overall gamma-ray background. This method was developed using a combination of established analytical techniques and photon transport simulations.

2.1. Gamma-ray background measurements

Six core-drilled concrete cylinders from three different composition concrete slabs were acquired from Oak Ridge National Laboratory (ORNL) for use during this research. The samples were labeled for identification. Samples from the same slab are identified with the same letter. The dimensions and masses of each sample were measured in order to calculate the density of the concrete. A picture of the concrete samples is shown in Figure 2.

Figure 2: The concrete samples used for this research.

Gamma spectra were collected for each sample using an HPGe semiconductor detector, the Genie™ 2000 spectroscopy software, and a multichannel analyzer (MCA). A lead vault was constructed around the detector to prevent interference from the concrete walls of the laboratory and radioactive check sources in the room. Prior to measuring any samples, a 24 h background spectrum was taken for later reference.After acquiring a background spectrum, a sample was placed in the vault and a 24 h spectrum was collected. This was repeated for the remaining samples. A picture of the detector geometry is given in Figure 3.

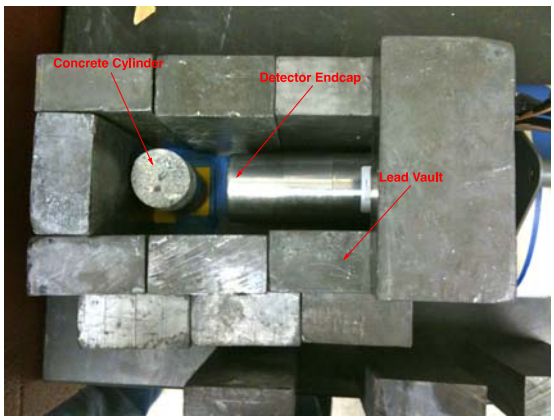

Figure 3: A picture of the detector geometry used for the concrete background measurements.

2.2. Neutron activation analysis

A combination of thermal and fast neutron activation analysis (NAA) was conducted to determine the elemental composition of the concrete samples. Prior to any analysis, gram-sized portions of each sample were broken off and ground into a powder.The powder was placed into polyethylene vials for irradiation. Additionally, each measurement involved the use of one or more comparator standards and quality control materials.

The neutron source for the thermal NAA irradiations was a 1 MW TRIGA research reactor located at the Texas A&M University Nuclear Science Center (NSC).The thermal neutron flux in the reactor was 10^{13} cm^{-2} s^{-1} during the experiments. For the first irradiation, the sample, standard, and quality control vials were sent into the core, irradiated for 30 s, returned, and allowed to decay for 1200 s. The sample vial was then placed on an HPGe detector and counted in live-time for 500 s.For the second irradiation, the vials were placed into an aluminum canister. The canister was sealed and irradiated in the reactor for 14 h. Afterwards, the vials were removed from the canister and allowed to decay for approximately six days, after which they were counted in live-time for 2000 s on anHPGe detector. After counting, the vials were placed aside and allowed to decay for an additional 20 days. Following the second decay period, the vials were counted in live-time for 10 800 s on the same HPGe detector.

The neutron source for the fast NAA irradiations was a Kaman Sciences Corporation A-711 sealed-tube neutron generator located at the Texas A&M University Center for Chemical Characterization and Analysis (CCCA). The A-711 generates 14.8 MeV fast neutrons through the $^{3}H(^{2}H,n)^{4}He$ fusion reaction.The beam intensity of the generator was between 10^{9} and 10^{10} s^{-1} during the measurements. The fast NAA measurements were used to determine the oxygen and silicon concentrations of the concrete. For the silicon measurements, the sample vials were sent to the generator, irradiated for 300 s, and returned. The samples decayed for approximately 60 s during transport to anHPGe detector where they were counted in live-time for 300 s.For the oxygen measurements, the samples were sent to the generator and irradiated for 20 s, then they were returned and counted on two summed NaI(Tl) detectors placed at 180º. The detectors were discriminated against any signals below 4500 keV. This method of oxygen determination with fast NAA was previously described by W. Ehmann and W. James [2,3].

2.3. Determination of concrete composition

The concentrations of multiple elements in the samples were determined using the relative method, which is given by:

$$\omega = \omega^{*} \frac{m^{*} A}{m A^{*}} \quad (1)$$

whereω is the concentration of the desired element, m is the mass of the sample, A is the decay-corrected activity of the sample, and the "*" superscript denotes the comparator standard [4]. The concentrations of carbon and hydrogen cannot be determined through thermal or fast NAA techniques; therefore, the concentrations of both elements were estimated for each sample based on the total weight percentage of the samples determined by the measurements. The total weight percentage of elements characterized through NAA for each sample was also determined by summing the individual concentrations.

2.4. Generation of efficiency calibration files with ISOCS™

After determining the composition of each sample, efficiency calibration files were generated using the ISOCS™ software. The measured dimensions and calculated densities were input into a vendor-supplied cylinder template. The NAA results were used to define a custom material for each concrete sample. A series of mathematical algorithms describing the response characteristics of the detector as a function of energy, angle, and distance from the detector was generated by the software for use with Genie™ 2000.

2.5. Determination of concrete background activity

Once the efficiency calibration files were generated, the background activities of the isotopes present in each concrete sample were calculated using the Genie™ 2000 software. The ISOCS™ calibration files were applied to each spectrum from the concrete samples. Gamma-ray energy peaks were located and their areas determined using the built-in algorithms available in the software package. The 24 h background spectrum was subtracted

from each sample spectrum, and nuclide identification was conducted using a customized nuclide library definition. The software was then used to calculate a specific activity for each of the gamma-ray energies found in the spectrum, and then compute a weighted mean specific activity for each identified isotope.

2.6. Generation of radioactive source terms for MCNP simulations

The background activity calculations were used to generate a source term for each sample that was used to define a distributed radioactive source throughout the concrete slab. The emission probabilities for theidentified gamma-ray peak energies in each slab were calculated as follows:

$$P_E^i = \frac{A^i y_E^i}{\sum_{i=1}^{N} A^i y_E^i} \quad (2)$$

where P_E^i is the emission probability of a gamma-ray with energy, E for an isotope, i, A^i is the total activity of isotope, i in the slab, y_E^i is the yield of a gamma-ray with energy, E, indicative of isotope, i, and N is the total number of identified isotopes in the sample.

2.7. MCNP simulations

The photon transport simulations were completed using the Monte Carlo N-Particle (MCNP) version 5 transport code [5]. A model of the physical RPM was constructed in MCNP and separate input files were saved for each sample. For each input deck, the NAA results were used to define the material composition of the concrete in the model. The radioactive source term calculations were used to create a distributed radioactive source throughout the volume of each concrete slab. An image of the RPM model is shown in Figure 4.

In order to determine count rates in the RPM, F8 pulse height tallies were applied to each PVT detector volume and the result was multiplied by the total gamma-ray emission rate for the concrete slab. Comparisons were then made between the count rate predicted by MCNP and the actual count rate recorded by the RPM in the presence of one of the concrete slabs. The model was validated by comparing the actual and predicted count rates for the RPM in the presence of varying-strength ^{137}Cs check sources.

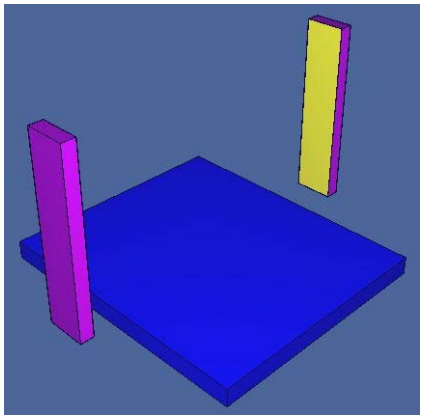

Figure 4: An image of the RPM modeled using the MCNP transport code.

Two sensitivity analyses were conducted to verify calculations and assumptions critical to the model. For the first analysis, the model was rerun with the concrete density set to ±2σ to determine the impact of large density fluctuations on the results. Since the carbon and hydrogen concentrations could not be determined with the NAA techniques employed, the second analysis focused on determining the impact of major fluctuations in their concentrations from an assumed value. Each deck was rerun with the carbon content set to 50% and 10% of this assumed value. The difference in the new total weight percentage and 100% was filled with hydrogen to prevent MCNP from renormalizing the weight fractions of the other elements in the slab.

3. Results and Discussion

3.1. Elemental composition of concrete samples

The concentrations of select elements in the quality control samples are given in Table 1 along with their associated literature values.The results show that for major constituent elements the quality control samples were within ±2σ of their stated literature values. Not only did this serve as validation of the comparator standards, but showed that the results for the concrete samples were both precise and accurate.The uranium and thorium concentrations of each concrete sample are given in Table 2. The results show a statistically significant difference in the uranium and thorium concentrations of samples G1 and G2 and the thorium concentrations of samples L1 and L2.

Element	Concentration ± σ	
	Quality Control	Literature [6]
Al	9.17 ± 0.12 %	9.18 ± 0.05 %
Fe	7.10 ± 0.01 %	7.21 ± 0.08 %
K	Below Limits	0.16 ± 0.01 %
Mg	1.18 ± 0.06 %	Below Limits
Mn	0.13 ± 0.01 %	0.12 ± 0.01 %
Na	1.53 ± 0.01 %	1.59 ± 0.03 %
O	30.77 ± 3.04 %	31.34 %
Si	28.35 ± 0.11 %	27.67 ± 0.27 %
Th	Below Limits	282 ± 19 ppb
Ti	0.77 ± 0.03 %	0.71 ± 0.02 %
U	Below Limits	320 ± 180 ppb

Table 1: Concentrations of select elements for quality control samples and their associated literature values.

Sample	Concentration ± σ (ppm)	
	U	Th
F1	1.16 ± 0.11	0.71 ± 0.22
F2	1.15 ± 0.07	0.94 ± 0.18
G1	2.75 ± 0.20	10.02 ± 0.05
G2	4.63 ± 0.32	14.26 ± 0.09
L1	1.54 ± 0.08	1.89 ± 0.02
L2	1.35 ± 0.09	2.25 ± 0.03

Table 2: Concentrations of uranium and thorium in different concrete samples.

3.2. Background activity of concrete samples

The specific activities of background isotopes in the concrete samples in given in Table 3. It should be noted that the ^{226}Ra and ^{232}Th isotopes are assumed to be in secular equilibrium with their respective daughter products. The ^{234}Th isotope – while the parent nuclide of ^{226}Ra – has been reported independently of the decay chain in order to validate this assumption.

Generally, between samples from the same slab, the specific activities of the identified isotopes are within ±2σ of each other. One exception to this is the ^{234}Th activity in samples from slab G. The calculated ^{234}Th activity in sample G2 is approximately 44% larger than the activity calculated for sample G1. From Table 3, the uranium and thorium concentrations of sample G2 are approximately 41% and 30% greater, respectively, than those of sample G1. Also, the specific activity of the ^{232}Th parent nuclide is approximately 7% larger in sample G1, indicating that although sample G2 has a greater overall concentration of thorium, sample G1 contains more of the naturally occurring ^{232}Th isotope. Since sample G1 contains a greater amount of ^{232}Th, and natural uranium is composed primarily of the ^{234}Th parent nuclide, ^{238}U, the greater specific activity of ^{234}Th in sample G2 is a result of the larger overall uranium content of the sample.

Sample	Specific Activity ± σ (Bq kg^{-1})			
	^{40}K	^{226}Ra	^{232}Th	^{234}Th
F1	42.27 ± 4.69	10.67 ± 0.50	4.97 ± 0.37	12.74 ± 2.28
F2	36.58 ± 4.63	10.55 ± 0.52	4.65 ± 0.38	13.34 ± 2.33
G1	696 ± 30	64.34 ± 1.84	80.21 ± 1.56	48.21 ± 5.15
G2	735 ± 32	67.70 ± 2.05	74.56 ± 1.56	85.56 ± 9.17
L1	170 ± 9	13.08 ± 0.61	9.58 ± 0.53	15.47 ± 2.50
L2	151 ± 8	12.70 ± 0.62	7.87 ± 0.46	18.43 ± 2.68

Table 3: Specific activities of background isotopes identified in different concrete samples.

3.3. Contribution of concrete to gamma-ray background

The average count rate per detector predicted by MCNP for each concrete sample is given in Table 4. Please note for this paper that the quoted uncertainties for MCNP results are only indicative of the MCNP statistical uncertainty.

Sample	Average Count Rate ± σ (counts s^{-1} detector^{-1})	
	0.0 – 3.0 MeV	40 – 140 keV
F1	84.29 ± 0.60	11.17 ± 0.22
F2	80.08 ± 0.58	10.71 ± 0.21
G1	950 ± 6	120 ± 2
G2	951 ± 6	120 ± 2
L1	164 ± 1	19.92 ± 0.39
L2	146 ± 1	17.83 ± 0.35

Table 4: The average detector count rate in the RPM predicted by MCNP.

The count rates determined by MCNP are consistent with the results from the specific activity measurements. Slab G had the highest total activity, followed by slabs L and F. It is expected that the larger activity would lead to a higher count rate in the RPM. It is worth noting that sample L1 has a count rate approximately 10% higher than that of sample

L2. The results given in Table 3 show that sample L1 has a total activity approximately 9% higher than that of sample L2. The higher total activity for sample L1 indicates that more gammas will be emitted from the slab and subsequently detected by the RPM.

A comparison of actual and estimated count rates from the RPM to the predicted values from MCNP for slab G is given inTable 5. The actual count rates for each detector were obtained by taking a 24 h average from a RPM data printout. It should be noted that the actual count rates from the RPM could also include counts derived from environmental backgroundor electronic noise. These extraneous signals make it difficult to determine the validity of the MCNP model, which only considers gamma-rays generated from the concrete slab. A 12 h average was taken from a RPM data printout for a time period where no concrete slab was present. This was subtracted from the actual count rates to obtain the estimated count rates.

Data Set	Count Rate ± σ (counts s^{-1})			
	Left, Upper	Left, Lower	Right, Upper	Right, Lower
RPM (Actual)	273 ± 17	315 ± 18	290 ± 17	338 ± 18
RPM (Estimated)	90 ± 10	144 ± 12	99 ± 10	161 ± 13
G1 (MCNP)	101 ± 2	137 ± 2	104 ± 2	137 ± 2
G2 (MCNP)	103 ± 2	140 ± 2	105 ± 2	134 ± 2

Table 5: A comparison of different RPM detector count rates for slab G.

In each case, the count rates in the upper detectors are less than those in the lower detectors. This is expected because the lower detectors are closer to the concrete slab, and therefore closer to the source of the gamma-rays. The data also shows that the MCNP results are consistent between samples G1 and G2. This is important to note because the larger amount of uranium and thorium in sample G2 did not impact the overall count rate. Since the highest yield gamma-rays from the decay of ^{234}Th are 63 and 93 keV, it is possible that they do not reach any of the PVT scintillators due to attenuation in the slab, the surrounding air, or the RPM structural material.

The data in Table 5 shows that it is possible to obtain a reasonable estimate of the gamma-ray contribution from the concrete using MCNP. Even though the estimated RPM count rates were within ±2σ of the MCNP results, there are a few sources of error that could impact the results. It was assumed that the lower level discriminator on the physical RPM was set between 40 and 140 keV. The count rates in the RPM could be higher or lower if incorrect discrimination settings were applied. Additionally, subtracting the 12 h averaged data from a different 24 h data set could also impact the results. Since environmental background can vary based on any number of external factors, the estimated RPM count rate may not be an accurate representation of the count rate from only the concrete.

3.4. Model validation and sensitivity analysis

The MCNP model was tested by measuring the response of the RPM to different strength ^{137}Cs check sources and comparing the count rates to those predicted by the code. A comparison of the RPM and MCNP count rates is given in Table 6. A ratio of the MCNP and RPM results was also calculated to identify and potential bias in the MCNP results. This ratio is also given in Table 6.

Source Strenth (μCi)	Count Rate ± σ (counts s^{-1})		MCNP RPM^{-1} ± σ
	RPM	MCNP	
5	18.75 ± 4.33	13.38 ± 0.14	0.71 ± 0.16
10	35.00 ± 5.92	26.77 ± 0.29	0.77 ± 0.13
15	50.60 ± 7.11	40.15 ± 0.43	0.79 ± 0.11
20	68.85 ± 8.30	53.53 ± 0.58	0.78 ± 0.09

Table 6: A comparison of RPM and MCNP count rates for various ^{137}Cs check sources.

The data in Table 6 shows that the count rate in the physical detector exceeded the count rate simulated in the MCNP model; however, the ratio of the simulated and measured data is statistically the same for all measurements. This validation case indicates that there is a certain amount of bias to the MCNP model. The RPM value was – on average – approximately 24% larger than the value predicted by MCNP. This difference could be caused by electronic noise introducing extraneous counts into the system. It is also possible that the energy window settings on

the RPM drifted during the measurements, causing an inconsistency with the MCNP simulations. Additionally, the MCNP model also does not incorporate the light collection efficiencies of the PVT scintillators or their photomultiplier tubes (PMTs).

Two sensitivity analyses were completed to verify measurements and assumptions in this research. Each MCNP deck was rerun with the concrete density set to ±2σ of the calculated value. A comparison of the average count rates is given in Table 7. Additionally, the MCNP decks were rerun with the carbon content set to 50% and 10% of the assumed value. A comparison of these results is given in Table 8.A carbon content sensitivity analysis was not completed for sample G2 because the elemental concentrations determined through NAA had already accounted for the entire weight percentage of the sample.

Sample	Average Count Rate ± σ (counts s^{-1} $detector^{-1}$)		
	-2σ	Calculated	+2σ
F1	11.26 ± 0.22	11.17 ± 0.22	11.14 ± 0.22
F2	10.76 ± 0.21	10.71 ± 0.21	10.63 ± 0.21
G1	121 ± 2	120 ± 2	120 ± 2
G2	121 ± 2	120 ± 2	120 ± 2
L1	20.03 ± 0.39	19.92 ± 0.39	19.80 ± 0.39
L2	17.97 ± 0.35	17.83 ± 0.35	17.76 ± 0.35

Table 7: A comparison of count rates for different concrete samples with varying densities.

Sample	Average Count Rate ± σ (counts s^{-1} $detector^{-1}$)		
	Assumed	50%	10%
F1	11.17 ± 0.22	10.82 ± 0.22	10.21 ± 0.21
F2	10.71 ± 0.21	10.44 ± 0.21	9.96 ± 0.21
G1	120 ± 2	119 ± 2	118 ± 2
L1	19.92 ± 0.39	19.53 ± 0.39	18.58 ± 0.38
L2	17.83 ± 0.39	17.82 ± 0.39	17.17 ± 0.34

Table 8: A comparison of count rates for different concrete samples with varying carbon content.

The data from Table 7 indicate that the density of the concrete will have a minor impact on the overall results. For this research, each case remained well within ±2σ statistical variation. Even though there was no adverse effect from these variations for this research, these simulations prove that the density of the concrete will have an impact on the number of gamma-rays reaching the detectors.

The data from Table 8 shows minor variations in the count rates as the carbon and hydrogen content of the samples widely fluctuated. For each sample, the 50% and 10% cases were within ±1σ and ±2σ, respectively, of the assumed concentrations. This indicates that an accurate estimate of the carbon and hydrogen content of a concrete sample can be made if the other elemental concentrations of the specimen are well known.

4. Conclusions and future work

4.1. Conclusions

Several conclusions may be drawn from the research presented in this paper. Primarily, the comparison of the actual and simulated count rates for slab G indicated that a quantitative estimate of the gamma-ray background contribution of concrete is achievable; however, other sources of radiation in the vicinity of the RPM need to be identified and characterized. The comparison of estimated and simulated RPM count rates from slab G indicates that the gamma-ray contribution of a particular concrete can be determined if the environmental background is well known.

The density sensitivity analysis showed that variations at ±2σ did not have a significant impact on the average count rate in the RPM detectors. It should be noted that the dimensions and mass of each sample were measured using digital instruments that had a high degree of precision. Low precision instruments will have an adverse impact on the uncertainties in the count rates.

The carbon and hydrogen concentration sensitivity analysis showed that the model is slightly sensitive to large variations in the concentrations of these elements. As long as the sample is well characterized by fast and thermal NAA, it is possible to make a reasonable estimate of the carbon and hydrogen content without the extra time and added costs of additional measurements.

4.2. Future work

There are a few areas where this work could be expanded in the future. It was assumed when comparing actual data to the MCNP results, that the RPM was correctly discriminated between 40 and 140 keV. The presence of a slight bias in the MCNP results indicates that this may not be true. If the physical detectors within the RPM employ an energy window larger than what was assumed, the count rate in the RPM would be greater than that predicted by the MCNP model.

In some instances the resources may not be available to conduct a full NAA characterization on a concrete sample. If concretes made with the same aggregate materials have slightly different elemental concentrations, but show comparable gamma-ray attenuation characteristics, it would be possible to use a standardized composition when modeling the material in ISOCS™ and MCNP. This would greatly reduced the time and costs involved in following this method.

Finally, the precision and accuracy of the model could be greatly improved by understanding the environmental background around the RPM. Taking multiple gamma-ray measurements near the RPM over a period of time would provide a data set that could be subtracted from the RPM count rates and provide a more accurate number for comparison with the count rates estimated with MCNP.

5. Acknowledgements

The authors would like to thank Dr. William James and Mr. Michael Raulerson of the Texas A&M University CCCA for the their assistance in carrying out the NAA work described in this paper. The authors would also like to thank the staff at the Texas A&M University NSC for allowing the use of their reactor facility to complete this research.

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Simulations of portal monitors to determine their applicability with respect to the detection of cargo-shielded radioactive substances

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Abstract:

To estimate the utilizability of radiation portal monitors (RPM) measurements under field conditions and simulations with MCNP were conducted. The best working detection conditions were determined studying the application of sodium iodide and plastic scintillators for an RPM. In the modeled scenario certain radionuclides (e.g. Cs-137) were located inside of cargo containers. The containers were inspected with an RPM. The radiation of the nuclides was attenuated by different types of cargo (3) filled in the containers. These cargo types were derived from statistics of US-American ports. Every cargo type is related to a certain amount of cargo containers imported and exported. The radiation background was simulated as well to determine the minimal detectable activities. The utilizability was evaluated regarding the measurement time and the number of used detectors in relation to a certain amount of sufficiently inspected containers.

Keywords: Gamma spectrometry, border control, MCNP

1. Introduction

The topic of this research project is the detection of illicit trafficking of radioactive material inside of cargo containers. The currently used radiation portal monitors (RPM) at border crossings are not sufficient due to two effects which often occur during operation: false positive and false negative alarms. In case of false positive an alarm is given in the absence of radioactive material (or natural and harmless radioactive substances are classified as relevant anthropogenic material). In the second case, false negative, radioactive material with high risk potential is not detected. Because of these two effects, more advanced detections systems and other technical efforts are desirable. The expected increase of cargo container flow and special security programs like the Megaports Initiative are demanding more reliable detection systems.

1.1. Main focus of research project

This research project focuses on the applicability of spectrometric gamma detection systems in general. Regardless whether currently used, advanced detectors or hypothetic gamma detectors, all need a certain amount of gamma radiation to identify characteristic peaks. The overall problem is that the emitted gamma radiation is attenuated and has to be discriminated from the radiation background. One has to consider that the attenuation of radiation depends on the energy of the gamma quanta, and that often in the lower part of the energetic spectrum a higher background occurs. This leads in particular to a larger decrease of detection probability of radionuclides in the lower part of the energy spectrum. One can assume that even with high ambitioned efforts only a small progress in detection efficiency will be achieved because the marginal improvements approach physical limitations that cannot be overcome.
The first part of the research project consisted of measurements with portal monitors with a low energy resolution and, in comparison, smaller spectrometric working detectors. This was done in order to get an overview about realistic measurement conditions, on frequently occurring radioactive substances

and on minimal detectable activities [1]. The second part was a computer modeling of portal monitors for gamma radiation. This was done to simulate gamma spectra of radionuclides inside cargo containers transported through a portal monitor.

2. Simulation

2.1. Modeled setup

The object of the simulation was to model a common cargo inspection for smuggled radioactive substances.

The examined scenario was modeled within an cube. Right in the center of the cube an RPM was located. The RPM was modeled with respect to 2 frequently used detector materials: sodium iodide and plastic scintillator. The RPM consisted in case of each detection material of four sub-detectors with equal volumes.

A homogeneous disk source was modeled below the RPM to simulate the natural radiation background. Concerning the radionuclides and their proportionate compounding, fix assumptions were made.

The inspected cargo container was considered as an unmoved object right in the middle of the RPM. In the center of the cargo container a radioactive point source was modeled. The radioactive gamma source inside of the container was assumed to emit gamma quanta in an isotropic way. To cover the different parts of the energetic gamma spectrum, gamma energies and emission probabilities of Ba-133 (low-energy), Cs-137 (middle-energy) und Co-60 (high-energy) were chosen. The source was surrounded by cargo.

In terms of the cargo, three different cargo types were developed and modeled. The attenuation of gamma sources inside of the container by the modeled cargo types was the main process of the simulation: The propagation of gamma quanta from a certain source through different shielding media to a detector. According to the physical qualities of the modeled cargo type, a certain amount of gamma quanta reached the detector and was able to establish an interaction which could then lead to a detection. Thus, the total detection efficiency of the examined measurement situation depends on the number of interacting gamma quanta. Since the number of gamma quanta that reaches the detector depends on the attenuating cargo, the total efficiency depends again on the cargo type.

Furthermore, different measuring conditions were evaluated considering the detection process itself (e.g. measurement time and the influence of the solid angle covered by the RPM).

2.2. Cargo classification and cargo types

At the Lawrence Livermore National Laboratory (LLNL) studies regarding the detection of radioactive sources by their emitted neutrons are conducted[2]. Like in the present study, the probability to detect a source inside of a cargo container is evaluated. The detection probability depends (like in the case of the detection of gamma quanta) on the shielding of the sources by cargo inside of the container. The shielding varies substantially by the physical qualities of the cargo, mainly by the atomic number (Z) and the material density (ρ). The LLNL-classification was developed with respect to the attenuation of neutrons. Since the present project deals with the attenuation of gamma quanta, the shielding potential of each cargo type had to be classified in a new way. This new classification was conducted regarding Z and ρ given in the LLNL-classification.

To determine the magnitude of shielding, the amount of gamma quanta was calculated for a certain shielding thickness. For this calculation the half width of a TEU container (117,5 cm) was chosen. By using the physical qualities of the LLNL-classification, the attenuation of gamma quanta was estimated at an energy of 662 keV (which is nearly the energy of gamma quanta emitted by a Cs-137 source). This estimation led to a categorization of the LLNL cargo types into 3 new cargo types with respect to their magnitude of gamma quanta attenuation. In table 1 the respective cargo type is presented as well as the related values for Z and ρ.

Cargo Type	Z	ρ [g/cm^3]
I	5	0,14
II	11	0,23
III	6	0,36

Table 1: Cargo types and their physical quantities in case of 662 keV. The atomic number Z and the density ρ are given for cargo type I (gamma transparent), II (gamma weak-transparent) and III (gamma non-transparent).

Each cargo type is correlated to a certain amount of container (given in TEU) that is necessary to transport (import and export) the classified cargo. In the present research project the correlation was conducted using data from the Statistical Office of Hamburg and Schleswig-Holstein.

3. Results of simulations

3.1. Minimal detectable activities

To determine the utilizability of an RPM it is useful to estimate the minimal detectable activity of a radioactive source. This leads to the concept of detection probability [3]. The goal of this concept is to determine the minimal given activity that can be detected with a certain probability. Therefore, it is necessary to evaluate the influence of the measurement time and the solid angle which is covered by the RPM. This leads to the necessary measurement efforts to detect a source with a certain probability. These measurement efforts depend on the total efficiency which itself depends on various measuring conditions, amongst others the shielding of the source. Since each shielding is correlated to a certain cargo type and at the same time to a certain amount of containers (TEU), it is possible to relate the necessary measurement efforts to a certain amount of containers (TEU).

Using this concept of minimal detectable activities the minimal measurement time was determined for a given detection scenario with a fix covered solid angle, depending itself on the total efficiency and the radiation background. Beside these physical quantities it was necessary to define error probabilities: In the discussed scenario the chosen probability for a type 1 error (false positive) was 0,14% and for a type 2 error (false negative) 5%.
The total efficiency of the detection process was used to determine the minimal measurement times obtained from the simulated spectra. In this context the total efficiency was calculated for a certain cargo type because the magnitude of the relevant photo peak compared to the rest of the spectrum depends on the attenuating modeled cargo. Furthermore, simulated background spectra were used to determine the count rate of the background.

Based on the two different ways of detector application – just measuring the gamma rate and gammaspectrometric measurement – the minimal necessary measurement times per cargo type were determined.

In order to examine a realistic scenario, the permitted limits for radioactive sources according to German law and the D-values (defined by the IAEA) were chosen to calculate the measurement times.

The minimal measurement times for the D-values (defined by the IAEA: Ba-133: 0,2 TBq, Cs-137: 0,1 TBq and Co-60: 0,03 TBq) were determined. For both detector types the minimal necessary measurement times were almost zero seconds for all three cargo types. Therefore, no further investigation was conducted with respect to the D-values.

The permitted limits according to German law are: 1 MBq in case of Ba-133, 10 kBq in case of Cs-137 and 100 kBq in case of Co-60. In case of the plastic scintillator detector the minimal measurement times were in the range of seconds for all cargo types. In case of the sodium iodide detector the minimal measurement times were in the range of hours to days.

The minimal measurement times calculated for the RPM based on plastic are presented in table 2 and the minimal measurement times in case of sodium iodide are given in table 3.

3.2. Amount of sufficiently inspected containers

Subsequently, the analyzed measurement times for the three cargo types were used to determine the quantity of potentially sufficient inspected containers handled in the port of Hamburg. For this purpose statistics regarding the transacted TEU in the port of Hamburg in 2008 and 2009 were evaluated.

Using the minimal necessary measurement time to detect radioactive sources inside of a container, it was possible to relate the respective measurement times per cargo type to a certain amount of cargo containers. For example, the minimal measurement time for the detection of a Ba-133 source inside of a container filled with cargo type I by the plastic scintillator detector amounts to 0,03 seconds. In the port of Hamburg approximately 11% of the imported containers are filled with cargo type I. Thus, for 11% of all containers imported in the port of Hamburg, a minimal measurement time of 0,03 seconds per container is required.

To ensure a reasonable number of sufficiently inspected TEU the reasoning is as follows:
The minimal measurement times necessary for the least shielding cargo type I, only allow a sufficient inspection for this cargo type. Containers transporting a more shielding cargo type are not sufficiently inspected. If, however, the minimal measurement time is given to inspect a container with a cargo type with higher attenuation capability, all containers transporting cargo with lower attenuation capabilities are inspected sufficiently as well.

Without a more detailed knowledge about the different cargo inside the containers, the only way to reach 100% of sufficiently inspected containers is to generally use the minimal measurement time analyzed for the most shielding cargo type III.

Ba-133 t_{min}[sec]	Cs-137 t_{min}[sec]	Co-60 t_{min}[sec]	Inspected Containers Import [%]	Inspected Containers Export [%]
1131	$3{,}0 \cdot 10^{6}$	$5{,}9 \cdot 10^{4}$	11,29	4,45
7553	$1{,}4 \cdot 10^{7}$	$1{,}9 \cdot 10^{5}$	50,34	58,77
208615	$2{,}2 \cdot 10^{8}$	$1{,}3 \cdot 10^{6}$	100	100

Table 2: Minimal measurement times in seconds per TEU for the sodium iodide detector. In the two right columns the percentage of sufficiently inspected imported or exported containers are given. The minimal measurement time, in case of the permitted statutory activity limits, is for a single container. The examined scenario deals with the container amounts handled in the port of Hamburg in 2009.

Ba-133 t_{min}[sec]	Cs-137 t_{min}[sec]	Co-60 t_{min}[sec]	Inspected Containers Import [%]	Inspected Containers Export [%]
0,03	102	0,87	11,29	4,45
0,1	230	1,44	50,34	58,77
1,09	1373	5,06	100	100

Table 3: Minimal measurement times in seconds per TEU for the sodium iodide detector. In the two right columns the percentage of sufficiently inspected imported or exported containers are given. The minimal measurement time, in case of the permitted statutory activity limits, is for a single container. The examined scenario deals with the container handled in the port of Hamburg in 2009.

4. Resume

The objective of this research project was to determine the utilizability of radiation portal monitors. For this purpose, portal monitors based on sodium iodide and plastic scintillator were studied. Analyzing a simulated detection scenario, the minimal measurement times for a sufficient container inspection to detect radioactive substances were elaborated. The minimal measurement times differed from each other depending especially on the respective cargo shielding of the radioactive sources. The minimal measurement times were determined with respect to the permitted limits for certain radionuclides according to German law.

Thereafter, the minimal measurement times were used to examine the possibility of an effective inspection of the containers handled in the port of Hamburg. Regarding the different types of cargo and their respective percentage in the entire container handling, it was analyzed how many containers might be sufficiently inspected within a reasonable period of time per container. For example, the necessary measuring time for the detection of a Ba-133 source inside of a container (filled with cargo type III) with a plastic scintillator detector was 1,09 seconds. On the other hand, the measuring time for the detection with a sodium iodide detector was more than two days.

Thus, the utilizability of spectrometric working detectors (like the sodium iodide detector) may only be affirmed in principle. In fact, in case of strong activities radionuclides can be detected within seconds but regarding weak activities (10 kBq to 1MBq) and eventually strongly shielding cargo, the necessary measuring time increases to hours or even days. Thus, the applicability of sodium iodide detectors for the purpose of identification of radionuclides remains problematic in the range of weak activities due to the low total efficiency of the detector.

Beside technical questions logistical problems occur: For example in case of just one RPM and a container handling of 6 million TEU in 2009 in the port of Hamburg, the maximum time for one container inspection would be about 5 seconds, measuring nonstop 24 hours a day. Therefore, the amount of RPMs would have to be increased significantly to ensure a smooth inspection of every container. But even if a higher number of RPMs avoided logistical defiles, an inacceptable high measurement time per container would remain.

In contrast to the sodium iodide detector, the much more sensitive plastic scintillator detector is also applicable in the case of weak activities. The minimal measurement time ranges within some seconds for activities in the range of the permitted limit. In case of strong activities (like in the case of the D-values) the plastic scintillator detector is applicable in any way. However, the applicability is limited, too, since the identification of radionuclides by their emitted characteristic spectra is not possible with a plastic scintillator detector.

5. Acknowledgements

The author would like to acknowledge Prof. Dr. Martin Kalinowski, Director of the Carl Friedrich von Weizsäcker-Centre for Science and Peace Research and Dr. Simon Labov, Director of the Radiation Detection Group at the Lawrence Livermore National Laboratory.

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Efficient use of low resolution personal radiation detectors at borders

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Abstract:

In 2008, the Finnish government decided to give funding to a project for upgrading the radiation monitoring systems at the Finnish border stations. The project is scheduled for the years 2009-2014, and the total funding will be 10 million euros. The aim of the project is to enhance the radiation monitoring operation at the Finnish borders so that the ability to detect the import of radioactive substances and nuclear material is improved.

In 2010, one hundred personal radiation detectors (PRDs) were purchased. The device is an easy-to-use low-price gamma and neutron detector. It has low resolution (CsI detector) spectrum acquisition and identification properties and it is capable of identifying a number of radioactive isotopes that are most typically encountered. The identified nuclides are classified as NORM, medical, industrial or Special Nuclear Materials. This is very useful for the customs officers, providing first hand information on the type of the detected radioactive material.

Automatic analysis of low resolution spectra is not always reliable. Therefore for efficient use of spectral detectors it is imperative that the spectra can be delivered to the experts for a more detailed analysis. For that purpose every customs station will be equipped with a mini-PC and software for downloading the spectra from the detectors. The downloaded spectrum file is in the LML (Linssi Markup Language) format, which can be uploaded to a central database via a secure web page. An expert can review the spectrum and perform an independent analysis, and provide expert support to the customs officer in almost real time. It is envisaged that the spectrum alone provides sufficient information so that correct conclusions can be drawn quickly, and unnecessary interruptions to shipments can be avoided.

In this paper the hardware, software and concept of operations are described.

Keywords: radiation; detection; spectrum; analysis; customs

1. Introduction

In 2008, the Finnish government decided to provide funding to a project for upgrading the radiation monitoring systems at the Finnish border stations. The project is scheduled for the years 2009-2014. The project is done in cooperation with Finnish Customs and Radiation and Nuclear Safety Authority, STUK [1].

In Finland Customs is responsible of monitoring the flow of goods across the borders. This includes radioactive substances and nuclear materials. STUK is an expert body providing Finnish Customs expert support (so called Technical Reachback[1]).

[1] *Technical Reachback* is the capability to assist the adjudication of radiation alarms or the discovery of suspicious or unauthorized materials that could be used to manufacture illicit radioactive or nuclear explosive devices that cannot be resolved

In 2010 the joint project plan included equipping border monitoring stations with altogether 100 handheld PRDs (Personal Radiation Detectors). The selected model is equipped with a CsI spectrometer and a 1024 channel MCA and has nuclide identification capability. The system is also equipped with neutron measurement capability. This handheld device will be one of the key tools for the Finnish Customs for years to come. Four different usage scenarios are envisaged. 1) Secondary examination after an alarm from another detector, 2) Continuous personal radiation monitoring, 3) Spectrometer facilitating Technical Reachback and 4) Ad hoc portal monitor.

2. Secondary examination device and continuous monitoring

Typically the device is used for secondary examination as a handheld device that Customs officers can carry along at all times. This is the most traditional way to use personal radiation detectors and therefore it is not discussed here. Customs personnel have been trained for this kind of use. The sensitivity of the small size detector is not very high but is sufficient in most cases. Additional neutron detection capability is an important feature. Spectral identification capability helps in some cases and may allow Customs Officers to conclusions independently.

3. Spectrometer facilitating technical reachback

As the device provides automatic identification of radioactive nuclides based on a low resolution spectrum, the analysis result is not always reliable. Also some other reasons may warrant for a more detailed examination of the measured spectrum, prompting the Customs officer to request expert support from STUK.

STUK has a 24/7 expert on duty system established for all kinds of nuclear and radiation emergencies. Through this system, Customs officers can reach STUK's experts for assistance. For those situations it is extremely useful that the Customs officer can send a spectrum to STUK for review. STUK has designed and set up a database for that purpose. The database is capable of receiving spectra measured in the field using various measurement systems, including the new PRD device.

In practice the Customs officers need to take the following steps. First, a spectrum is measured and stored in the memory of the PRD. Then it is downloaded to a PC with dedicated software. Finally, the spectrum is uploaded to the central database via a secure website.

The goal of the cooperation between Finnish Customs and STUK is to improve the Technical Reachback that STUK provides to the Customs. At the moment Customs officers receive Technical Reachback support by phone and fax. This practice has many drawbacks: The lack of detectors with spectrometric capability makes it difficult to perform identification and to assess the situation. Oral information transfer from the border to the Technical Reachback at STUK is sometimes ambiguous, lacking in accuracy and specificity. The communication may be biased by personal perceptions and is therefore not accurate or reliable. Information may also be incomplete. Thus, the final assessment of the case is often based on inaccurate or incomplete information.

Some of the deficiencies above will be addressed by the use of new PRDs and the renewed concept of operations. If a Customs officer needs support, he/she can call the expert on duty by phone. When the measurement is taken in the field, the spectral data can be stored in the database where it can be immediately reached by Technical Reachback. With modern encryption technologies this data can be made available via the Internet in a secured manner. The concept of operations is shown in figure 1.

at the detection site. Technical Reachback relies heavily on radiation analysts and subject matter experts who can identify specific isotopes and potential threats based on data collected from the detection site, either remotely, or in person.

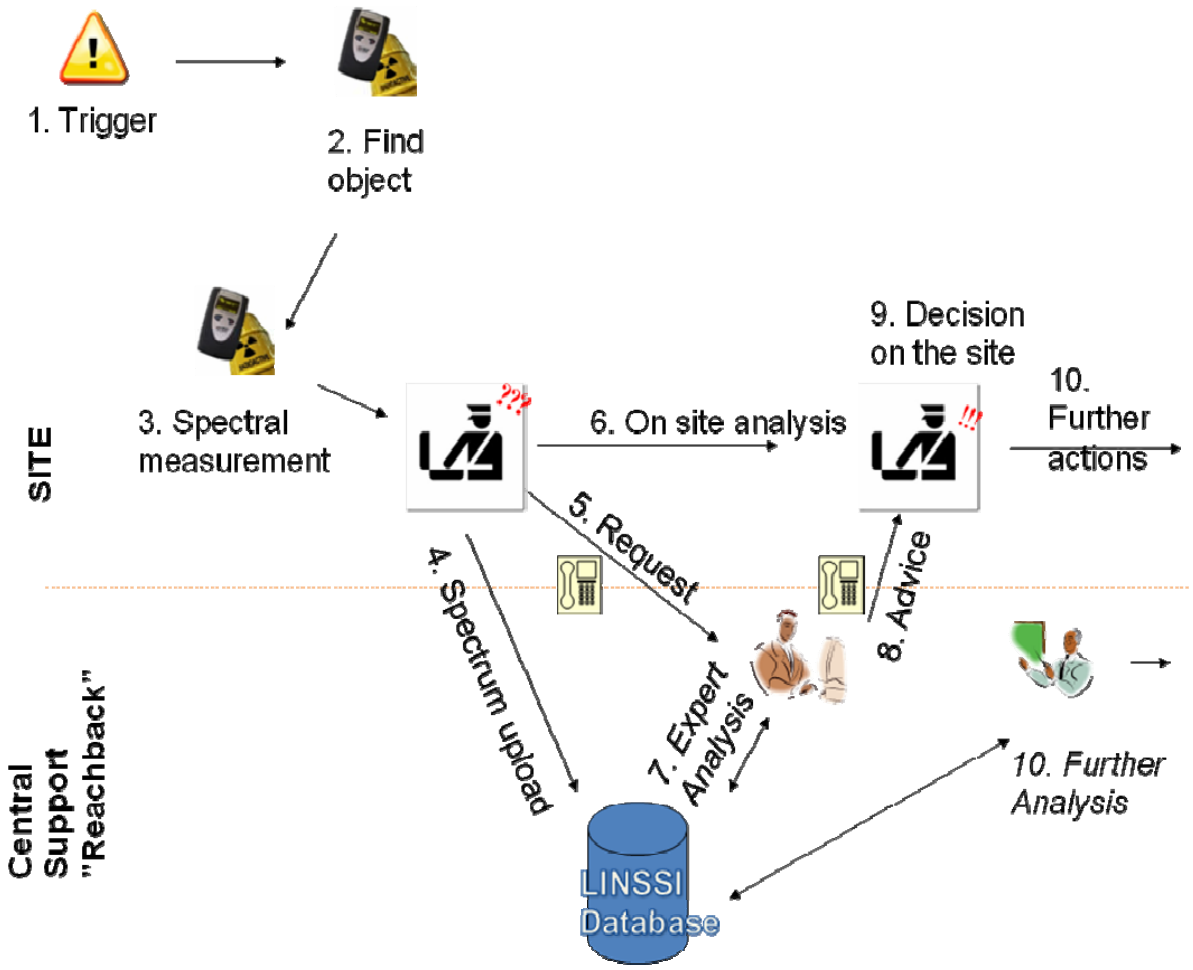

Figure 1: Concept of operations in case of PRD detection.
1. Trigger (for instance an alarm from a non-spectral portal or a continuously operated PRD).
2. The object is stopped for inspection and localized.
3. Secondary spectral measurement with PRD.
4-5. Customs officer decides to request for Technical Reachback. He/she uploads the spectrum to LINSSI database and calls to STUK's expert on duty.
6. On site analysis and assessment continues.
7. Review and analysis of the data by the expert.
8. Advice from expert to the Customs Officer in charge of decision.
9. Decision by the Customs Officer based on on-site and expert analyses.
10. Subsequent actions and analyses may follow.

4. Ad hoc portal monitor

If the PRDs are equipped with rechargeable batteries and a charger connected to the grid, it can operate continuously over long periods of time. The PRD can be connected to a small laptop or mini-PC via a USB cable; for this purpose STUK has designed a wall bracket which can hold both a PRD and a laptop. The software used to download spectra is also capable of displaying real time measuring data and announcing alarms both in numerical and graphical format. This kind of measuring system, when installed in the customs officer's booth, is able to give an alert when a source above the detection limit passes by. The detection limit depends on geometry and passing speed, but sources which elevate gamma dose rate for more than 0,2 µSv/h should be readily detected. PRDs in continuous mode will also complement traditional portal and pillar monitors, providing immediate secondary information. If alarms from both a portal monitor and a PRD go off simultaneously, one can be sure that something radioactive is approaching. The worst case scenario is that extremely highly radioactive and hazardous sources are passing the border station. In those cases sensitive portal monitors may be saturated and lower detection efficiency of PRDs is a desired feature.

5. LINSSI database functionalities

Central database needed to support the Concept of Operations presented in the previous structure exists and is in operational use. LINSSI is an SQL database for gamma-ray spectrometry [2]. It has been developed in collaboration with the Radiation Physics Group of Helsinki University of Technology, STUK, and the Radiation Protection Bureau, Health Canada. LINSSI is freely available for all registered users or for users who let the developers know that they have installed the software. Users may modify the database tables to achieve the functionality they desire.

LINSSI database was originally created to store and control successive analyses of gamma spectrometric data created by CTBT radionuclide monitoring network. LINSSI has many applications today, varying from gamma spectrometric laboratory measurement analyses to environmental monitoring performed at stationary real-time networks. Moreover, LINSSI is also in use with mobile units and in nuclear safeguards applications. LINSSI is in operational use in the external radiation monitoring network of Finland maintained by STUK.

LINSSI database is at its strongest in storing gamma spectrometric measurement data, supporting multiple analyses on the same dataset and creating multiple views to the same data. However, other types of data can also be stored. LINSSI is Linux OS based and can be installed in portable and industrial stand-alone PCs.

The user group of LINSSI has created a multitude of useful scripts and reports that can be used for border monitoring applications. Measurement data can be automatically uploaded to the database, if it is in the supported XML-type format (LML, Linssi Markup Language format). LINSSI also supports automatic gamma spectrometric analysis pipeline routines originally written for CTBT applications. These routines are successfully applied in mobile environmental measurements, which do not have specific measurement geometries. LINSSI reports are available remotely, providing analysis tools for Technical Reachback experts. In addition, successive analyses of given data can be performed online. Therefore, LINSSI can act as a seamless interface between the original data creator, automatic pipeline analysis and Technical Reachback.

6. Conclusion

STUK has procured for Finnish Customs portable Personal Radiation Detectors (PRDs). It is now the most common radiation detection tool in Finland, since all Customs offices in Finland will have it at their disposal. The device has new features like spectrometric identification capability and a continuous measurement mode, which are useful for the work of the Customs. Spectral information from the detectors will be transferred to STUKs expert, who will provide Customs officers with Technical Reachback.

7. Legal matters

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11 Safeguards by Design

Safeguards by Design (SBD) for the Next Generation Safeguards Initiative (NGSI)—Accomplishments and Future Plans

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Abstract:

The International Atomic Energy Agency (IAEA) has described the safeguards-by-design (SBD) concept as an approach in which "international safeguards are fully integrated into the design process of a new nuclear facility from the initial planning through design, construction, operation, and decommissioning." International safeguards features often are added following completion of the facility design. Earlier consideration of safeguards features could reduce the need for costly retrofits of the facility and could result in a more efficient and effective safeguards design. The United States National Nuclear Security Administration (NNSA) is sponsoring a project through its Next Generation Safeguards Initiative (NGSI) to promote a global norm for the use of SBD for international safeguards. The NGSI SBD program has been developed in parallel with a similar effort at the IAEA while taking into account the IAEA's SBD achievements and future plans. The NGSI program includes DOE laboratory studies, international workshops, engagement with industry and the IAEA, and the setting of an example through its planned use in new nuclear facilities in the US. Consistent with this effort, the NGSI program has sponsored "lessons-learned" studies, the preparation of facility-specific SBD guidance documents, and an SBD workshop at the third international meeting for NGSI (NGS3). The following paper describes past accomplishments and future plans for the NGSI SBD program.

Keywords: Safeguards; Design; NGSI

1. Introduction

The United States (US) National Nuclear Security Administration's (NNSA's) Office of Nonproliferation and International Security (NA-24) recently began a project through its Next Generation Safeguards Initiative (NGSI) to establish a global norm for the use of safeguards by design (SBD) for international safeguards. More specifically, this effort is focused on establishing a global norm for the incorporation of safeguards features early in the design phase of a new nuclear facility to avoid the need to redesign the facility at a later date or retrofit the completed facility. In doing so, not only can redesign or retrofitting be avoided, but consideration of safeguards features early in the facility design activity can provide a more efficient and effective implementation of International Atomic Energy Agency (IAEA) safeguards. This efficiency and effectiveness can be gained by (1) relying on proven safeguards design concepts based on lessons learned and (2) optimizing facility features that influence safeguards implementation but are not typically affected by safeguards requirements, such as a plant's layout. The NGSI program is being developed in parallel with a similar effort at the IAEA while taking into account the IAEA's SBD achievements and future plans.

NGSI SBD activities currently include lessons-learned studies, the preparation of "model" SBD guidance documents, industry engagement to aid in SBD guidance document preparation and to gain acceptance for the SBD concept, and collaboration with the IAEA toward preparing internationally accepted SBD guidance. The NGSI SBD guidance documents are referred to as "models" because NA-24 hopes they will form the basis for internationally recognized guidance, in some cases drafted by the IAEA Member States in support of the IAEA Secretariat and its SBD program. NGSI SBD guidance documents prepared to date are based on NNSA studies that have identified best practices for satisfying IAEA safeguards objectives and advanced concepts that may enhance future safeguards performance. Best practices have been determined from lessons learned in operating facilities, and advanced concepts consist of innovative instrumentation and novel design features. Preparation of the model SBD guidance documents has benefited from early engagement with industry by providing insight into designers' and operators' needs.

An example of best practices for uranium enrichment facilities is the provision for IAEA access to the UF_6 header pipe connecting the feed station and cascade hall. This provision for access facilitates design verification and the installation of accountancy instrumentation, such as cascade header enrichment monitors. IAEA access to the feed header has generally not been a feature in past enrichment facility designs and cannot be added easily after facility construction (if at all). An example of a novel design feature for a uranium enrichment facility is a window in the feed autoclave that allows an inspector to verify the presence of a feed cylinder visually and perhaps read the cylinder identification plate. The first of these examples, the UF_6 header, may affect the location of walls and piping and therefore requires consideration early in the design process. The second of these examples, the autoclave window, would require early consideration by the manufacturer to make autoclave design changes and would possibly require testing to demonstrate safety.

Close collaboration with the IAEA on SBD guidance documents will be important for achieving the long-term goal of establishing a global norm. Once complete, the guidance documents will provide industry an opportunity to achieve increased safeguards effectiveness at potentially less expense to the IAEA and facility operator.

2. Discussion

Although numerous stakeholders have an interest in SBD, the primary objective of the NGSI program to date has been to develop model SBD guidance for the nuclear facility designer. The longer-range objective, to convert the NGSI model SBD guidance to internationally accepted SBD guidance, will be accomplished through international workshops and conferences, as well as industry and IAEA engagement. By describing best practices and advanced concepts for IAEA safeguards approaches, the facility designers will be provided example safeguards features from the outset of the facility design activity that have a high likelihood of satisfying IAEA safeguards objectives. Other stakeholders with an interest in SBD include the facility operator, domestic regulatory agencies, and the IAEA.

To date, the NGSI SBD studies have included

- nonfacility specific general guidance for SBD,
- SBD lessons learned from existing facilities,
- success criteria for safeguards design,
- safeguards evaluations for "trade studies",
- "safety by design" as a template for SBD,
- implementation of the SBD process,
- model SBD guidance.

The NGSI program has sponsored the preparation of a number of model SBD guidance documents to date including documents for Gas Centrifuge Enrichment Plant (GCEP) uranium enrichment facilities, Gen III/III$^+$ light-water reactors (LWRs), and the US Department of Energy's (DOE's) Next Generation Nuclear Plant (NGNP) gas reactor. In addition to the preparation of model SBD guidance documents, NGSI SBD

activities have included industry engagement with those currently planning, designing, or building new uranium enrichment facilities in the US and those interested in advanced fuel recycle facilities, to include reprocessing and reactor technologies. The discussions with industry have included sharing preliminary SBD guidance based on IAEA inspector experience and DOE laboratory research and development and gathering feedback from industry representatives regarding the information they believe to be relevant for SBD. NNSA engagement with the uranium enrichment industry has been coordinated with the US Nuclear Regulatory Commission (NRC) as part of ongoing licensing discussions.

2.1. Specific accomplishments

2.1.1. NGSI SBD guidance-related documents

More specifically, the NGSI SBD studies have resulted in the following publications. A general nonfacility-specific SBD guidance document was prepared by Thomas [1] that describes in broad terms how IAEA safeguards are implemented and provides examples of high-level IAEA safeguards objectives. Identification of best practices through lessons-learned studies of existing facilities include the Thorp study by Hebditch et al. [2] and the Rokhasho Reprocessing Plant study by Johnson and Ehinger [3]. Other NGSI studies include the identification of success criteria for safeguards design by DeMuth and Scherer [4] and a review of methodologies for estimating proliferation resistance as a tool for trade studies by Mullen and Scherer [5]. The study by Mullen and Scherer highlights two recent international efforts to develop a methodology for assessing proliferation resistance that can then be used in trade studies to make safeguards effectiveness comparisons (i.e., trades) for competing technologies and/or facility designs. These two international efforts are (1) the International Project on Innovative Nuclear Reactors and Fuel Cycles (INPRO) and (2) the evaluation methodology for Proliferation Resistance and Physical Protection (PR&PP) of Generation IV nuclear energy systems. Such efforts could result in facility designs that make diversion or misuse more technically difficult and easier to detect. Another NGSI-sponsored SBD task has been adapting the “pathway analysis” approach and several of the analytical measures developed in the PR&PP methodology to support a Facility Safeguardability Analysis (FSA) methodology. The objective for an FSA methodology is to provide the facility design team with a tool to assess the “safeguardability” of a proposed facility’s design as early as the preconceptual design stage. FSA should aid facility designers in identifying safeguards issues early in the design process and provide them with a framework for selecting facility-specific SBD practices and lessons-learned to resolve these issues. Additionally, FSA should help the designer anticipate how innovations in their designs might pose new safeguards issues that could be addressed through changes in the design, or enhancing elements of the safeguards approach in a manner likely to meet IAEA safeguards objectives, while mitigating cost impacts on both the IAEA and facility operator (see Durst et al. [6]).

Model SBD guidance documents for uranium enrichment prepared by Durst et al. [7] and updated by Whitaker et al. [8] were shared with the IAEA for its feedback. Other model SBD guidance documents include that for Gen III/III+ LWRs prepared by Pan et al. [9] and the US Next Generation Nuclear Plant (NGNP) prepared by Durst et al. [10, 11]. In addition to the preparation of model SBD guidance, an NGSI study was initiated by Bjornard et al. [12] to identify approaches for implementing SBD. In this context, implementing refers to the use of project management principles for application of SBD. The study by Bjornard et al. is intended to provide examples of good project management practices for industry. It was the view of the authors that highlighting good project management practices can lead to more careful planning related to the timing of integrating safeguards features into the overall facility design activity. And finally, to gain additional insight for SBD guidance, a study by Hockert et al was commissioned to ascertain lessons learned from the development and implementation of safety by design. [13]. Safety by design became a subject of interest several decades ago that led to numerous methodologies for determining the effectiveness of safety measures.

2.1.2. Industry engagement for new US plants

The US Nuclear Regulatory Commission (NRC) has recently hosted efforts to facilitate the use of SBD for new uranium enrichment facilities currently being planned for construction in the US (see Grice et al. [14]). Although SBD is not an NRC requirement, the NRC is aiding the implementation of SBD by coordinating

discussions between DOE's Office of Nonproliferation and International Security (NA-24) and the industry's facility design teams. These discussions have included Louisiana Enrichment Services (LES) with respect to its GCEP facility under construction in Eunice, New Mexico; AREVA with respect to its planned GCEP facility in Eagle Rock, Idaho; and Global Laser Enrichment (GLE) with respect to a laser enrichment facility under consideration.

NGSI industry engagement has also occurred with respect to future plutonium recycle facilities in the US. Preliminary discussions related to SBD for surplus plutonium recycle from U.S. defense programs have been conducted with Shaw AREVA MOX Services, LLC, who will be the operator of the Mixed Oxide (MOX) Fuel Fabrication Facility currently being built at the DOE Savannah River site in Aiken, South Carolina. SBD discussions related to the recycling of plutonium from spent fuel have been conducted with AREVA and General Electric (GE). Discussions with AREVA have focused on potential future plutonium recycle in the US for reactor-grade plutonium. This process may be based on advanced aqueous separations such as AREVA's co-extraction (COEX) process, with potential modification for actinide separations, initially recycling plutonium in LWRs with conventional MOX fuel and later in fast reactors to include the minor actinides. Discussions with GE were focused on plutonium separation by electrochemical processing, to be recycled with or without minor actinides in fast reactors. Additionally, efforts have been initiated to discuss SBD with General Atomics (GA) regarding its NGNP.

2.1.3. International workshops and symposia

In October 2008, the IAEA hosted an international workshop to discuss how safeguards could be better facilitated in nuclear plant design and operations. The outcomes of the meeting were published in the document "Facility Design and Plant Operation Features That Facilitate the Implementation of IAEA Safeguards" (STR 360) [15]. One of the key recommendations of that document was that the IAEA continue engaging all stakeholders in the SBD process and creating expert working groups on, among other things, SBD principles based on facility type. During the annual Institute of Nuclear Materials Management (INMM) symposium July 11–16, 2010, in Baltimore, Maryland, the NGSI program sponsored a session on SBD during which 10 papers were presented by representatives of governments and industry. In November 2010, the NGSI sponsored several SBD posters at the IAEA safeguards symposium in Vienna (for example see DeMuth and Lockwood [16]).

The *Third International Meeting on Next Generation Safeguards*, hosted by DOE/NNSA's Office of Nonproliferation and International Security (NA-24) on December 14–15, 2010, at the Washington Hilton Hotel in Washington, DC, was a 2-day technical meeting to discuss implementation of the SBD concept. There were approximately 100 meeting participants from thirteen countries, comprised of safeguards policy and technical experts from government and industry. Also present were representatives from the Brazilian-Argentine Agency for Accounting and Control of Nuclear Materials (ABACC), the European Atomic Energy Agency (EURATOM), and the IAEA. The primary objective of this meeting was to exchange views and provide recommendations on implementation of the SBD concept. Through written working group reports, these perspectives could help form the basis for internationally endorsed guidance documents that outline the application of SBD for four different nuclear fuel cycle facility types: enrichment, reactors, reprocessing, and fuel fabrication [17].

2.2 Future Plans

In 2011 the NGSI will sponsor the preparation of three new model SBD guidance documents prepared by the DOE national laboratories. These documents were selected by considering the interests of the IAEA and US DOE. The new SBD documents will be focused on (1) uranium conversion, (2) research reactors and (3) interim spent fuel storage installations. In 2011 and beyond, the NGSI will continue engaging industry for SBD of new nuclear facilities in the US. As in 2010, the NGSI will sponsor a session on SBD at the INMM annual meeting planned for July 17–21, 2011, in Desert Springs, California. And finally, the IAEA, with assistance from the European Commission Support Programme, is currently developing a document that will describe the basic principles of safeguards and the fundamental design features and measures that facilitate the implementation of international safeguards. The IAEA has also discussed plans to propose a support program task to the participating IAEA Member State Support Programs to

prepare facility-specific SBD guidance relative to those facility types in which specific States have experience and expertise. Consequently, it is expected that the IAEA efforts to further SBD will be facilitated by the accomplishments to date, and future activities of, the NGSI program.

3. Conclusions

The NGSI SBD program was initiated in 2008 to develop standards for safeguards design at an early phase in the overall facility design activity. Studies initiated for the NGSI effort were related to lessons learned from implementation of safeguards in existing facilities, identification of best practices and advanced concepts for safeguards design, approaches to trade studies for evaluating safeguards design options, and implementation of SBD in new nuclear facilities. The NGSI SBD program has progressed from the concepts phase to that of defining safeguards approaches that have a high likelihood of meeting IAEA objectives. The method selected by the NGSI team for enabling SBD at the industry level is the development of "model" SBD guidance documents. It is expected that NGSI SBD guidance can serve as a model for preparation of internationally accepted SBD guidance, in some cases drafted by the IAEA Member States in support of the IAEA Secretariat and its SBD program. The NGSI is pursuing its goal of making SBD the global norm through industry engagement, international workshops, and setting-an-example by encouraging the use of SBD in new nuclear facilities in the US.

4. Acknowledgment

This work has been sponsored by the US DOE/NNSA, Nonproliferation and International Security (NA-24), NGSI.

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A Safeguardability Check-List for Safeguards by Design

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Abstract

Safeguards by design is a complex step-by-step interactive decision process involving various stake-holders and design choices to be taken over a certain period of time.
The resulting design should be a compromise among economical, safety, security and safeguards implementation aspects.

Access to technology and equipment, as well as to the nuclear fuel cycle, determines the basic choices that the designer has to make. Once the boundary conditions for a given facility have been fixed, the designer still faces the challenge of setting several design and operational parameters that will require various trade-offs . These can be seen in three groups, i.e. those related to the general design and its intrinsic proliferation resistance; those related to the specific lay-out and planning; those related to the actual safeguards instrumentation, its effectiveness and efficiency.

The paper aims at describing a model for a phased, or "layered" approach to safeguards-by-design, focusing on the example of off-load reactors.

Keywords: Safeguards by Design, SBD, safeguardability, nuclear installation, safeguards, proliferation resistance

1. Introduction

The final design of a nuclear facility is the result of a compromise optimizing the purpose and intrinsic features of the facility (scope, process, materials, planning) with economic, operational, safety and security factors, by taking into account the safeguards needs at an early design stage.

Safeguards by design (SBD) is a complex multi-disciplinary step-by-step interactive decision process involving various stake-holders and design choices to be taken over a certain period of time. The goal of the SBD approach is to fully integrate safeguards into the design process of a nuclear facility, from the initial planning through design, construction, operation, and decommissioning. Taking into account the safeguards needs since very early design stages would be beneficial for all the involved stakeholders

The IAEA started a dedicated activity in October 2008 [1]. High level guidelines are being finalised by IAEA based on EURATOM Support Programme's input [2]; facility specific guidelines are under preparation. They will be particularly important for developmental facilities with new design features and technologies that require R&D and new SG approaches.

2. Safeguards approaches

All the EU civil nuclear installations are under EURATOM safeguards. The legal framework under which the EURATOM Safeguards are enforced is given by Commission Regulation (EURATOM) n. 302/2005. All operators of civilian nuclear installations have to comply with the provisions laid down in such document, which also regulates the information needed to be submitted in case of construction of a new nuclear installation. Safeguards by design are not legally required to date, and could only be implemented on a voluntary basis.

Following the entry into force of the Additional Protocol in many countries, the IAEA aims to have "integrated safeguards" in force. In EU this applies since 2010.

Integrated safeguards are based, *inter alia*, on various types of short-notice, unannounced and complementary access inspections, which are aimed at providing assurance that no undeclared nuclear activity is carried inside or outside declared nuclear sites, as well as the data provided by the "classical" international safeguards system components:

- Design information verification (DIV);
- Nuclear material verification;
- Containment and surveillance (C/S) measures

In the broader context of fully-information-driven safeguards, IAEA's analyses and conclusions on the absence of proliferation activities are drawn by assessing the whole State's potential, and not only the nuclear material accountancy at facility level, which however remains the basis of the safeguards system.

The footprint of the nuclear facilities installed and declared in the country should in the end be compatible with the official declarations.

In this respect, the analyses take into account also a series of indicators of non-classical indicators like environmental monitoring, satellite imagery, open source information and trade analysis.
Indeed the AP also requires states to declare exports of "trigger list" items possibly assisting the establishment of 15 nuclear fuel cycle related activities, listed in AP Annex I.
Figure 1 shows a pictorial view of this approach.

3. The Safeguards by Design process

The safeguards-by-design process, as described e.g. in [3], should be conceived mainly looking at the classical components with a direct influence on safeguards friendliness, but also on the indirect impact on non-classical indicators. A design choice with its associated input and output generates a series of emanations which should also be reflected in the analysis. This means considering e.g. imports of technology, equipment, fresh fuel, need for maintenance and spare parts, ancillary dual use equipment etc, as well as satellite imagery and environmental analysis.

The decision to build a new facility is strategic, economic and political. It's typically taken by the government, approved by the Parliament and tasked by the Regulator to an identified energy operator. Various boundary conditions apply. The new facility is part of the country's fuel cycle (NFC), and can serve to replace an old, or even a missing element of the NFC. The decision should take into account the country's access to technology, equipment and resources import, as well as its international commitments.

The operator launches a call for tender to which suppliers and manufacturers answer with proposed designs and budget. The Regulator will verify the compliance with safety standards, as well as security and safeguards requirements [4].

The weighing factors differ subjectively, saving the relative high importance of safety and costs. Nevertheless, the regulator and operator must be aware of the needed technology fixes and costs associated with the proliferation resistance "intrinsic design features", and in order to fulfil the "extrinsic measures", i.e. safeguards objectives. The less "safeguards-friendly" the design is, the higher is the amount and cost of the safeguards system needed to ensure the fulfilment of objectives.

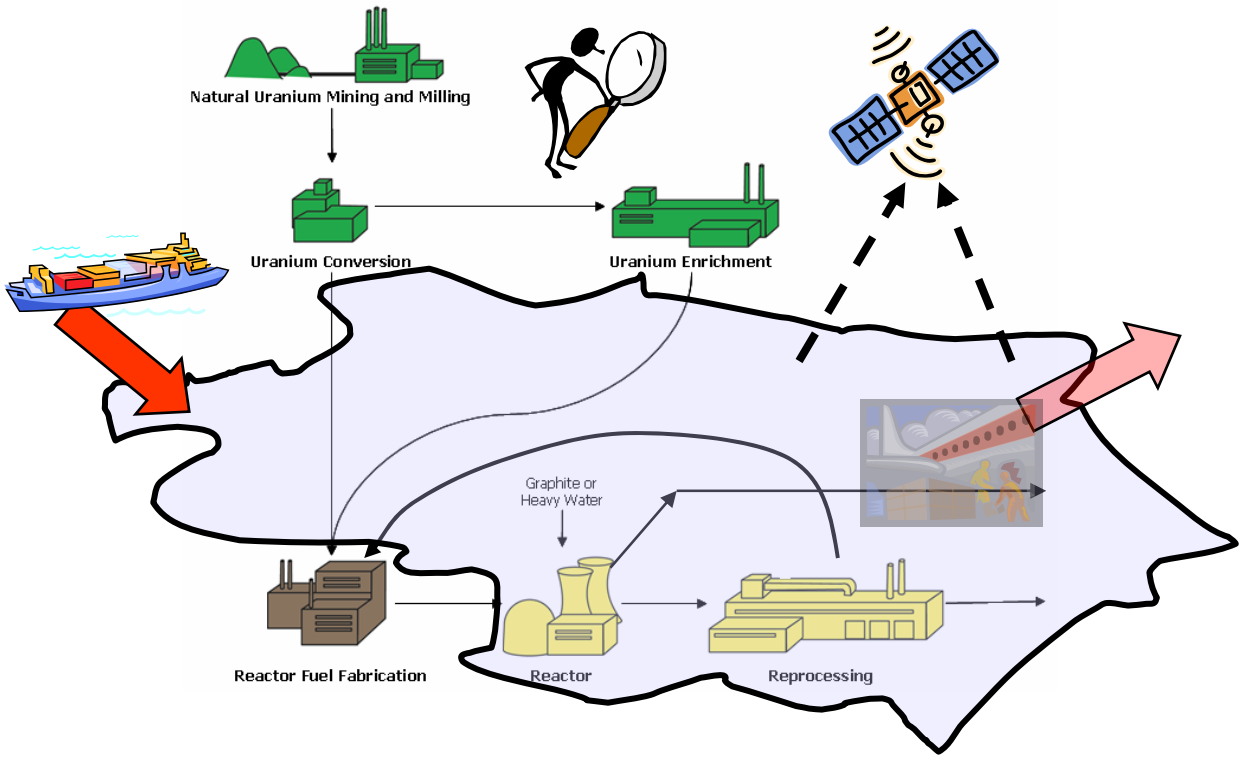

Fig. 1 - Fully information-driven safeguards

The resulting compromise design is in the end a trade off among the various needs within the budget, ensuring the desired production (e.g. enriched uranium, fuel elements, energy).

A comprehensive and interacting SBD process should hence already begin during the R&D phase, with an exchange of information between the safeguards inspectorates, the national authorities, the operator and the designer:

The safeguards inspectorates should provide designers with a statement of safeguards requirements as early as possible when plans for a new facility are communicated.

The national authorities, in turn, should provide the safeguards inspectorates with preliminary design information as soon as they are made available by the supplier through the tendering procedure launched by the operator.

Safeguards guidelines specific for the facility type in object, should be available to the designer, for a first concept to be presented to the operator, the national authorities and to safeguards inspectorates.

Low-level details and requirements should then be addressed at the beginning of the facility design and construction process, specifying the safeguards system performance and test acceptance criteria. As a result, the development of the safeguards approaches and their elements should ideally match the new facility's milestones, as summarised in Tab.1.

4. Safeguardability

Safeguardability is defined as "a concept that reflects the degree of ease with which a facility can be put under safeguards" [5] and is an approach which can be used in the conceptual and preliminary design phases.

It consists in a list of safeguardability attributes, i.e. intrinsic design features with safeguards relevance, which should be taken into account by system designers, guiding them through design choices and decisions.
An initial set of safeguardability attributes is listed in [5, 6], and a more detailed list included in [7]. The scope of the present paper is to propose a non-exhaustive extension of the safeguardability tables, with the additional idea to try and organise the attributes also according to their relevance to each particular design phase, as well as to the safeguards components.

Tab.1 summarises the milestones and decision steps following the government's decision to build a facility of certain general characteristics and output. Each step implies safeguardability consequences and R&D needs.

Phases	R/SSAC	IAEA	Operator	Designer
R&D phase	Provide general information on new facility build to IAEA	Best safeguards practices compilation for the facility type		
	Identifies operator		Call for tenders to designers/suppliers	Facility pre-concept tenders
			Tender selection	
	Tender assessment against safety, security and safeguards			
Pre-Conceptual design				Preliminary design concept
	Approval		Approval	
	Preliminary design information to IAEA	Safeguards requirements; High level SG guidelines		
Preliminary Design				Preliminary design
	Approval		Approval	
	DIQ	Medium level SG guidelines		
		Safeguards approach		
		Detailed SG guidelines		
		Design information evaluation		
		Feed-back to R/SSAC		
	Feed-back to operator		Feed-back to designer	Feed-back to safeguards equipment supplier
Final design	Draft facility attachment			
Construction		DIV; Safeguards installation	Safeguards installation	
Commissioning	Final facility attachment	DIV; Safeguards testing	Safeguards testing; Possible feed-back to equipment supplier	Possible feed-back to equipment supplier
Operation	Accompany inspections; Provide info to IAEA under AP	Inspections; DIV	Comply with inspections; Provide info to SSAC for AP declarations	

Tab 1: Safeguards by Design process

5. A phased approach to SBD: the Safeguardability Check-List

The attributes are grouped in three categories corresponding to the classical safeguards system components, but also in three layers according to the impact they have on the design phase and

development, with the available level of details. Each layer corresponds to one or more of the design phases of Table 1. In each layer, each feature may affect one or more safeguards components.

First layer: design basic facility's features intrinsic to the process

It includes design features intrinsic to the process with direct proliferation resistance and safeguardability relevance. Their definition relates to R&D and pre-conceptual design phase of Tab. 1.

The basic parameters are given by the type of facility, its main characteristics and desired output, i.e. electricity production for a reactor. These are an input to the safeguardability analysis, linked to the consequent decision on fuel type.

Again in the reactor's case, the choice of the fuel type and fuel cycle has an economic justification, and varying safeguards implications. It is guided by the associated needs of enrichment, production, transport, storage, core design, type of loading, irradiation, maximum burn-up, post-irradiation handling and reprocessing and final storage requirements.

Each design choice has an impact on safeguardability, because the attractiveness of the nuclear material varies along the fuel cycle, and also the tracing/monitoring/measurement of the nuclear material changes, requiring different types of techniques and instrumentation, with varying performances.

Second layer: design features related to facility's lay-out and safeguards installation

A second layer of design features is linked to the actual planning and lay-out of the facility, and definition of the safeguards model. Previous similar designs can serve as first model, as well as existing safeguards approaches.

The interaction between the designer, the operator, the regulator and the safeguards inspectorates is crucial in this phase, which poses the main foundations of the design and can avoid the need for costly retrofitting.

Typical choices relate to foreseeing cabling, Key-Measurement Points (KMP), Physical Inventory Taking (PIT), access for inspections, transfer routes, remote automated handling.

An eye should be of course kept on the instrumentation to be installed later and its requirements, also in relation to remote monitoring. Interaction with instrumentation experts and R&D is therefore mandatory.

During planning and lay-out, care should be taken to facilitate transparency of design, visual surveillance and inspection, use of 3D laser reconstruction for DIV and equivalent techniques.

Third layer: Design Features related to safeguards efficiency and effectiveness

Once the first two layers are defined, the stake-holders have to choose and install the safeguards equipment. The nuclear material's level of radioactivity, or signature, chosen in the first layer influences the choice of the detectors' type and their efficiency and effectiveness.
Shared-use equipment and remote data transmission should also be taken into account.

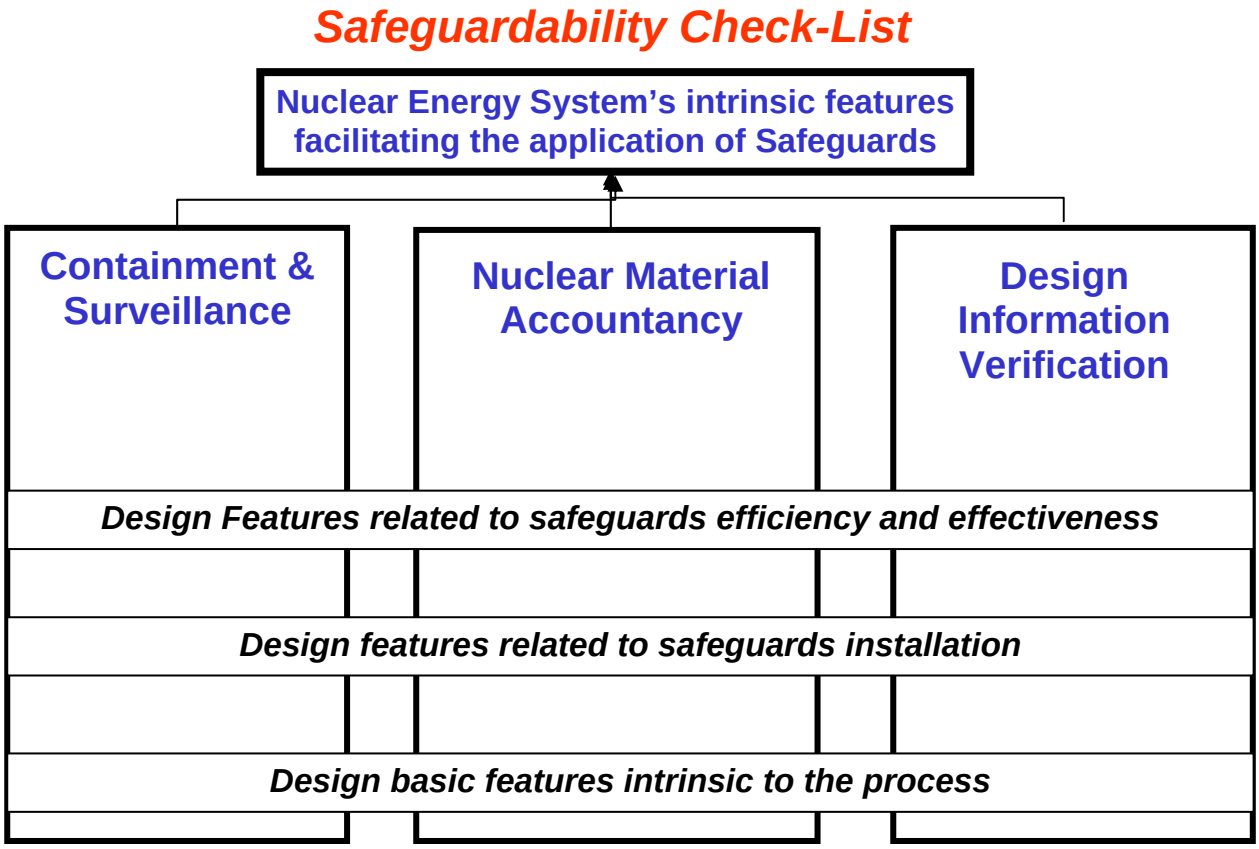

Fig. 2 - The Safeguardability Check-List model

6. Discussion

The proposed list of attributes is presented in Appendix. For ease of reading the features are tabled according to their safeguards components main relevance, though they may influence also another.

Each attribute has a suggested metrics with values in the range between 0 – 1, proportional to the estimated safeguardability relevance. The higher is the value the higher is the safeguardability.

> *Example: at increasing level of automation, safeguardability and possibility of remote monitoring increase*

On the right end columns, for each attribute it is also indicated to which of the three *layer*(s) of the design each attribute belongs. In this way, it is easy either to sort the attributes per layer or per safeguards component.

The safeguardability relevance of the features (e.g. extent of access to the core, or type and burn-up of the fuel) can be estimated by means of Utility functions. Where available, they are referenced to. Missing ones can be estimated by expert elicitation.

Following the check-list, one can interactively assign safeguardability value to each attribute based on expert judgment or estimates by available utility functions.

After filling the whole table the designer can see where the safeguardability is strong and where it has weaknesses. The best value for each attribute is higlighted to give the designer an idea what he should look for. Of course some of the attributes can not be changed some of them are fixed resulting from other decisions. Typical examples of such attributes are power of the reactor or NM throughput.

The list of attributes presented relies a lot on previous studies, as referenced. It could be expanded and tailored to different types of facilities. The example presented refers to reactors, and it illustrates how the metrics and estimates of each safeguardability attributes could be presented.
Each attribute can impact more than one safeguards component, and intervene at different design stages, primary secondary

No aggregation is proposed in this phase of the study; nevertheless there appears to be dependence among various attributes which should be further investigated. Aggregation towards a final "safeguardability" score could be useful to rank the overall ease of implementing safeguards, but is not straightforward and would require the investigation of several parameters not obvious to set up.
An overall view of the model, either in tabular form as in Appendix, or as decision tree can help to better visualise the areas requiring intervention, maintaining a view of the details at lower level.

7. Conclusions and way forward

The work presented in this paper describes the basis of the check-list approach and the role of time dependence. The intention was to try and shape a modular phased analysis supporting safeguards by design. The list of suggested attributes is open for comments and suggestions. It can be expanded or modified. At the same time, attribute metrics will be improved, and where applicable more utility functions can be introduced or created.

To facilitate the use of SCL, one could pre-calculate all relevant attributes for different types of nuclear materials circulating in the facility (fuel types, front-end, back-end materials), in order to establish datasets that could be easily used for qualitative parametric studies by stake-holders.

The next proposed step is then to verify the relevance of the proposed attributes and metrics with an SDB demo study, supported by simulation of the safeguards system efficiency and effectiveness.

A second level of development of the safeguardability check-list may wish to identify and asses the interactions among attributes for a more sophisticated fine tuning of the model.

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APPENDIX: Safeguardability Check-List attributes

1. Nuclear Material Throughput

Each facility has a nominal NM throughput. This factor is measured in [SQ/year] and evaluates the nuclear material flow. The bigger the flow is the bigger the safeguards effort is needed. Significant quantity is defined as the approximate amount of nuclear material for which the possibility of manufacturing a nuclear explosive device cannot be excluded. [6].

2. Nuclear Material Average Inventory

Each facility will contain some inventory of NM. The inventory is measured in significant quantities. The smaller the inventory is the less safeguards effort is needed. Nuclear material inventory in the facility can vary through time of facility operation. For purposes of estimating safeguardability the average inventory should be used.

3. Nuclear Material Attractiveness [FOM]

Nuclear material attractiveness may vary along the facility. The value can be calculated along the whole fuel cycle, and for the benefit of the facility design in some topical conditions along the transfer route. Some key values could be e.g. for fresh fuel storage, core fuel (because it differs with the length of irradiation – the value in the middle of irradiation should be used), spent fuel (upon discharge) and fuel going to interim storage.
This type of assessment may highlight the more “attractive” spots, or else the ones where more safeguard effort is needed and with a shorter time for diversion detection.

NM’s attractiveness can be estimated e.g. by the Figure of Merit (FOM) provided by LANL in [8]:

$$FOM_1 = 1 - \log_{10}\left(\frac{M}{800} + \frac{Mh}{4500} + \frac{M}{50}\left[\frac{D}{500}\right]^{\frac{1}{\log_{10} 2}}\right)$$

Where *M* is the bare critical mass of the metal in kg, *h* is the heat content in W/kg, and *D* is the dose rate of *0.2·M* evaluated at 1 m from the surface in rad/h.

4. Weight fraction of even plutonium isotopes

Even isotopes cause higher neutron rates and heat generation rate. Both of those factors can be used for passive detection of NM and make higher proliferation resistance of NM. This attribute differs along the facility as the NM attractiveness.

Weight fraction of even plutonium isotopes is discussed in [9] and [10]

5. Concentration of NM

Higher concentration materials will be more attractive since a lower mass (or volume) of material would need to be diverted or stolen to acquire a useable mass of SNM. The metric uses the number of SQ’s of material per metric ton (1000 kg) as its input value. The SQ definitions of the IAEA are used (i.e., 8 kg for Pu, 25 kg for HEU, 75 kg for LEU, 25 kg for Np-237, 25 kg for Am, and 20000 kg for Th). The maximum value of concentration is 125 SQ/Mt for Pu. Usually there will be more type of material in the facility so weighted average should be used for concentration assessment.

Inspired by [9].

6. Radiation signature

Radiation signature determines the easiness of identifying/recognizing the type, composition and amount of nuclear material. For verification of nuclear material destructive analysis and/or passive or active non-destructive NDA analysis can be used. Passive NDA, based on spontaneous emissions of

neutrons, gamma rays or the total heat decay, are in general quicker and less source demanding then the DA and active NDA and thus preferred.

Originally inspired by [11] p. 13-14.

7. Frequency of outages

Frequency of outages determines the frequency of the core opening. Every core opening means some movements around spent fuel pond and requires bigger safeguards effort or inspectors' arrival. Also reliability of outages should be taken into account. If the outages are going according to the scheduled inspection planning it's much easier.

8. Fraction of processes treated as items

In terms of nuclear material accounting items are easier and cheaper to verify than NM which is treated as bulk [6].

9. Radiation field for direct inspections

Key Measurements Points (KMPs) should be designed in such a way that radiation field should be as small as possible. The maximum radiation field in this safeguardability analysis was set as a dose rate in which the inspector would work for 180 days, 8 hours a day to receive a dose 20 mSv which is usual annual dose limit for radiation workers.

10. Accessibility of NM for PIV

PIV is usually an annual activity which is performed upon closing of NM balance when NM Inventory is verified by item counting attributes testing and/or other possible methods and this inventory is compared to declared inventory by the operator. In certain cases NM can be placed in such a place which for certain reasons can not be accessed by an inspector. NM Inventory is determined by NM flow from/to this area. In such cases reliable NM flow monitoring must be employed to keep continuity of knowledge. Some remote verification equipment could be used.

11. Near real time accountancy applicable

For some processes the closure of material inventory for timeliness purposes is particularly challenging. Having the possibility to implement near real time accounting would greatly help the inspectorate in closing the material balance frequently and without interrupting the process, in order to achieve timeliness objectives while not intruding in the system's operation [6].

12. Possibility to use shared equipment

Using of shared equipment with the operator can facilitate obtaining data and safe resources of acquitting, maintaining and operating safeguards equipment. But concerns about integrity and authenticity of data can occur [2].

13. Measurement uncertainty

In bulk handling facilities the differences between book inventory and physical inventory can occur due to measurement uncertainty and the nature of the process. Those uncertainties should be as small as possible to deter diversion hidden behind measurement uncertainties [12].

14. Items dismountability

NMA in item handling facilities is widely based on item counting and serial numbers cross-check. If there is a possibility to item disassembly more inspection effort has to be performed to detect potential diversion of rods or pins.

15. Items labelling

Labelling should be reliably difficult to remove, replace or falsify. It should also possibly be verifiable in an automated way (e.g. cross-checking identification (ID) info by means of laser scanning surface image analysis, and radiation dose signals).
This would be beneficial to a more efficient application of safeguards and would help also to detect misuse or diversion scenarios implying replacement of items with dummies [2].

16. Water quality in spent fuel pools

Cerenkov viewing device is widely used practice how to verify content of spent fuel pool. The other methods are more time consuming and intrusive. Water quality is important factor determining if CCVD is possible to use or not [4].

17. ID tag can be read without moving the item

Being able to read the ID tag without having to move the item would result in a large saving of time and therefore of efficiency, especially in situations where a large number of items are to be checked. This property would turn to be useful also for the operator in relation to his internal accounting purposes [7].

18. ID tag legible after the whole process

The designer should take into account all processes which the item will have to undergo and should design the labeling in such a way that the ID will be legible after all activities [7**Error! Reference source not found.**].

19. NM Holdup

In a nuclear material bulk handling facility there will be locations and areas where there will be a build up of nuclear material arising from the processing operations. These locations need to be clearly identified and the estimated quantities expected calculated. The fluctuation of NM holdup should be as small as possible [2].

20. Homogeneity

For bulk handling facilities homogeneity can significantly contribute to measurement uncertainties because the amount of NM is often determined from small sample. Or if the material is measured with gamma spectrometry the samples should be homogeneous because gamma rays are absorbed differently in different densities [7].

21. Personnel accessibility

The assessment of personnel accessibility was taken from [9]. The scale was chosen to reflect a decrease in proliferation resistance as the difficulty in accessing the material decreases. “Inaccessible” implies that the material cannot be physically accessed (for instance material being irradiated in a PWR). A “canyon” refers to a completely enclosed, underground structure to which it is very difficult to gain access. A “vault” refers to a large structure that impedes access to the material (a spent fuel pool was considered a vault in this work). “Secure” refer to sealed containers in which material may be stored (this could include drums or barrels). “Remote” would refer to any system in which its location alone makes it inaccessible to the proliferator (a geological repository is typically one example of this). “Hands-on” refers to engineered configurations in which the material can be at least indirectly handled (i.e. very limited physical barriers, such as a glove-box).

22. Limited number of possible transfer routes

Increasing number of possible transfer routes boosts C/S effort.
[6]

23. Extent of automation

Appropriate degree of automation enables inspectors recording and analyzing movements of important devices like fuel handling machines etc.
[6]

24. Standardization of fuel transfer

Standardization of fuel transfer in terms or transfer routes and transport containers facilitates application of C/S. For example it makes easier interpretation of recorded images easier surveillance of standard items in transit as they would probably result in moving at the same speed.
[6]

25. Layout facilitating MBA and KMP definition

The facility should be laid out so that its physical structure and boundaries can be mapped onto MBAs and that KMPs can be easily determined. The safeguards inspectors should ensure that they are satisfied with their integrity during the construction phases.
[2]

26. Hidden access to nuclear material

A system's area might have numerous openings and connecting routes needed for a large variety of activities as e.g. maintenance activities, or personnel access to the area. Having the possibility of determining that all these openings could not be used as transfer routes for diverting nuclear material would allow to optimise the surveillance activities, providing additional coverage to the most sensitive areas.
[7]

27. Autonomous lightening and electricity supply for C/S cameras

For maintaining continuity of knowledge it is important to have autonomous electricity supply and lightening for C/S cameras. The cabling and space for these instruments should be taken into account in design stages.

28. Power supply reliable and redundant

The illumination system in areas under optical surveillance should be considered as a critical system, and its availability guaranteed. This includes also the assessment that no interruption of power supply could reasonably affect the illumination system.
[7]

29. Intensity of illumination is adequate

Together with illumination dynamic range, the illumination intensity is an important parameter for assessing the performance that a surveillance camera might achieve in a given situation. If illumination is too low the camera sensor might be challenged to capture information at very high sensitivity settings, losing details and increasing the image noise. A too strong illumination might "blind" the camera, preventing it to record the activities that are going on. The latter situation could occur during particular activities requiring very strong temporary light sources: If these are inadvertently (or intentionally) pointed towards the camera lenses, the camera would be momentarily blinded and continuity of knowledge might be lost.
[5, 6]

30. Homogenous illumination

Even with a modern ccd or cmos, situations in which the illumination is not homogeneous could result in a situation where the high contrast could prevent the camera to retain the necessary amount of detail over the whole picture. Having a constant and homogeneous illumination would ensure that the cameras are always in their optimal working conditions.
[5, 6]

31. Casual switch-off

When interruption of illumination is concerned, one of the causes might be the inadvertent switching off of the lights by operators. Having an illumination system that prevents this eventuality would help to rule out one of the most probable causes of illumination interruptions.
[5, 6]

32. Radiation level compatible with C/S equipment

Surveillance equipment positioned in processing areas where high radiation levels exist might experience various degrees of interferences, spanning from interference with the recording sensor to the impossibility of e.g. sending the recorded data to a remote server outside the radioactive area wirelessly. Although radiation hardening of the equipment is possible and generally implemented, taking into account the interaction between the process activities and the surveillance ones would help to increase the overall efficiency of the safeguarding activities. In addition, the opportunity to have a less harsh working environment might give the inspectorate the possibility of applying less radiation hardening and therefore of using less expensive equipment. This opportunity might also be taken by the operator when designing his physical protection system.
[5, 6]

33. Clear field of view for the C/S cameras

Placing of C/S cameras has to be designed during design stages due to allocate cabling and electricity supply. However, the camera views are best confirmed after construction phase when all the construction and commissioning scaffolding and other obstructions have been removed. This can partially be overcome using computer simulation models. While designing the C/S placing also operational practices should be taken into account.
[5, 6]

34. Seals applicable

The inspectors should take advantage of physical barriers which enable application of seals. These physical barriers should be inspected during construction phase. In the design stages usage of proper hooks for seal's rope should be taken into account. For transport of NM standardized flasks optimized for sealing should be used.
[2]

35. Remote data transmission

Remote monitoring enables reducing the need of inspectors to physically visit the facility.
Inspector visits take up travel time on the IAEA side and impact on the operation of the facility. If the need to visit can be reduced, this would lessen the impact on the operator. This, however, expands the use of instrumentation.

Data should be consolidated at the facility in a centralised location, preferably outside the controlled area. Often times, sensor data need to be retrieved at multiple locations throughout the facility. If data collection can be centralised, preferably outside controlled areas, time spent at the facility could be reduced.
[2]

36. Labelling of equipment

All safeguard equipment including cabling should be clearly labelled.
[5, 6]

37. Comprehensiveness of documentation

Every facility put under international safeguards will have to be described for safeguards purposes. For Design Information Examination (DIE) and Design Information Verification (DIV), a Design Information Questionnaire (DIQ) has to be compiled. Exact and complete documentation in both hardcopy and electronic form would facilitate DIQ compilation.
[5, 6]

38. Inspectors' access during construction phase

It would be beneficial if inspectors had access to the facility during construction phase, were treated as staff and were provided by offices and equipment to make their conclusion and analyses.
[13]

39. Transparency of layout

During DIV process equipments and layout are verified. Especially in bulk handling facilities with complicated structures transparency of layout might help to detect modification of the process in order to diversion or undeclared production of NM.
[5, 6]

40. Possibility to use 3D laser scanner

3D laser based scene detection is state of the art technology used for Basic Technical Characteristic/ Design Information verification. It increases the probability to detect design modification then naked eye or photos comparison. Designer should make sure that those techniques can be used as easily as possibly by communication with experts and taking into account that those equipments are quite big and heavy.
[5, 6]

41. Accessibility during operation

The system should be conceived in such a way that every relevant process equipment can be visually or instrumentally checked for DIV purposes during the facility normal activity.
[5, 6]

Appendix B: Safeguardability Check-List application to nuclear reactor safeguards design

Facility data

					Features intrinsically linked to PR	Features related to safeguards installation	Features related to safeguards efficiency and effectiveness
Facility class	existing	evolutionary	innovative				
SG approach exists	yes	no					
Fuel flow	continuous	in batches	items				
Fuel type							
Physical state of NM	solid	liquid	gas				
U-235 enrichment (%):							
Pu-enrichment (%):							
U-233 enrichment (%):							
Burn-up (GWd/MT)							
Number of MBAs							
Number of KMPs							

NM Accountancy

1	NM throughput [SQ/year]	0 >100	0,3 50-100	0,6 1-50	1 <1				x
2	NM average inventory [SQ]	0 >100	0,25 40-100	0,5 30-40	0,75 1-30	1 <1			x
3	NM Attractiveness [FOM]		FOM	0 >2	0,3 1-2	0,6 0-1	1 <0		x
		Fresh fuel							
		Core fuel							
		Spent fuel							
		Interim storage							
4	Weight fraction of even Pu isotopes		0 0-0.2	0,25 0.2-0.4	0,5 0.4-0.6	0,75 0.6-0.8	1 0.8-1		x
		Fresh fuel							
		Core fuel							
		Spent fuel							
		Interim storage							

NM Accountancy

							Features intrinsically linked to PR	Features related to safeguards installation	Features related to safeguards efficiency and effectiveness
5	Average concentration of NM [SQ/Mt]	0 100-125	0,3 70-100	0,6 20-70	1 0-20		x		
6	Radiation signature	Fresh fuel Core fuel Spent fuel Interim storage	0 Material with no reliable signature allowing for remote detection	0,3 Materials requiring active and/or intrusive means of detection, but with moderate detection probabilities	0,6 Materials which can be reliable detected but require active means	1 Materials easily detected by passive means with unique radiation signatures that are difficult to shield	x		
7	Frequency of outages [months]	0 0-12	0,5 12-24	1 >24					x
8	Fraction of NM processed as items	0 0-0.2	0,25 0.2-0.4	0,5 0.4-0.6	0,75 0.6-0.8	1 0.8-1			x
9	Radiation field for direct inspections [µSv/h]	0 >112	0,25 84-112	0,5 56-84	0,75 28-56	1 <28			x
10	Accessibility of NM for PIV	0 Neither visual inspection nor NDA verification possible	0,3 No visual inspection but NDA possible with medium detection probability	0,6 No visual inspection but NDA possible with high detection probability	1 Visual inspection and NDA analysis or samples taking possible				x

								Features intrinsically linked to PR	Features related to safeguards installation	Features related to safeguards efficiency and effectiveness
NM Accountancy										
11	Possibility to implement near real time accountancy	0 No	1 Yes							x
12	Possibility to use shared equipment	0 Shared equipment not possible. No attempt in design stage to introduce shared equipment.		0,5 Partial introduction of shared equipment		1 Using shared equipment as wide as possible. Negotiated with IAEA in early design stages. Effective and reliable authentication.				x
13	Expected measurement uncertainty (relative standard deviation)	0 >0.25	0,2 0.188-0.25	0,4 0.126-0.188	0,6 0.064-0.126	0,8 0.002-0.064	1 <0.002			x
Facility using items:										
14	Items dismountable	0 Disassembly is allowed and part of "routine" practice		0,5 Disassembly is not allowed in design but is easy and difficult to detect		1 Difficult to disassemble and easily detectable			x	
15	Items labeling	0 Labeling less reliable than the current practice		0,5 Current practice labeling		1 Labeling more difficult to falsify then current practice			x	
16	Water quality in spent fuel pool	0 Insufficient water quality for using CCVD		1 Water quality is sufficient for using CCVD						x

NM Accountancy

					Features intrinsically linked to PR	Features related to safeguards installation	Features related to safeguards efficiency and effectiveness
	Facility using items:						
17	ID tag can be read without moving the item	0 No	1 Yes				x
18	ID tag legible after the whole process	0 No	1 Yes				x
	Bulk handling facility						
19	NM Holdup	0 NM holdup not clearly identified with big fluctuations and uncertainties.		1 Holdup as small as possible. Optimized design. Locations clearly identified and holdup estimated.		x	
20	Homogeneity	0 Highly inhomogeneous materials like waste, scrap etc. Special measurement methods have to be introduced	0,5 Inhomogeneities contribute to uncertainty of measurements	1 Homogeneous materials			x

Containment and Surveillance

								Features intrinsically linked to PR	Features related to safeguards installation	Features related to safeguards efficiency and effectiveness
21	Personnel accessibility	0 Hands-on	0,25 Remote	0,5 Secure	0,75 Vault	0,9 Canyon	1 Inaccessible			x
22	Number of transfer routes is limited	0 >7	0,2 6	0,4 5	0,6 4	0,8 3	1 <2	x		
23	Extent of automation	0 Manual operation		0,5 Partial automation		1 Full automation with possibility of recording movements and further analysis.				x
24	Standardization of item transfer	0 Transfer of items is chaotic and causes problems for transfer monitoring		0,5 Medium level of standardization		1 Items are transferred at constant velocity. Routes are optimized for monitoring.				x
25	Layout facilitating MBA and KMP definition	0 Not reliable physical barriers of KMP and MBA.		0,5 MBA and KMP possible to be defined and monitored but with minor changes of facility.		1 KMP/MBA definition. Layout discussed with IAEA. Integrity of barriers verified during construction phase.			x	
26	Hidden access to nuclear material	0 yes	1 no							x
27	Autonomous lightening and electricity supply for C/S cameras	0 no	1 yes						x	
28	Power supply reliable and redundant	0 no	1 yes						x	

Containment and Surveillance

						Features intrinsically linked to PR	Features related to safeguards installation	Features related to safeguards efficiency and effectiveness
29	Intensity of illumination is adequate	0 no	1 yes				x	
30	Homogenous illumination	0 no	1 yes				x	
31	Casual switch-off possible	0 yes	1 no				x	
32	Radiation level compatible with C/S equipment	0 no	1 yes				x	
33	Clear field of view for the C/S cameras	0 Field of view often limited		0,5 Field of view is limited during the limited number of processes	1 Field of view is always clear. Processes optimized for optical surveillance		x	
34	Seals applicable	0 Seals not easily applicable		0,5 Seals applicable with minor changes of design	1 Layout is optimized for seal application. Proper hooks are used. Physical barriers are checked for integrity.		x	
35	Remote monitoring possible	0 Design not optimized for remote monitoring		0,5 Remote monitoring possible after minor changes.	1 Data are gathered to centralised place outside the controlled zone. Reliable connection and data flow.			x

Design Information Verification

						Features intrinsically linked to PR	Features related to safeguards installation	Features related to safeguards efficiency and effectiveness
36	Labeling of equipment	0 Lack of labeling causes difficulties and impede the inspectors' effort		1 All safeguards equipment and cabeling is clearly labeled.			x	
37	Comprehensiveness of documentation	0 Incomplete documentation which requires further supplentation and causes delays of completing DIQ		0,5 Supplementary questions for clarifying documentation necessary	1 Exact and complete documentation about the facility. Both the hardcopy and electronic form.			x
38	Access for inspectors during the construction phase	0 No access to inspectors during CP		0,5 Limited access for inspectors. Long administrative procedures upon arrival.	1 Inspectors treated as staff during the CP. Provision of officess and equipment.			x
39	Layout is transparent	0 <10%	0,3 30% transparent	0,6 60% transparent	1 100%		x	
40	Possibility to use 3D laser scanner	0 No	0,3 At around 30% of processes	0,6 At around 60% of processes	1 Can be used wherever			x
41	Accessibility during operation for DIV	0 No	1 Yes					x

New nuclear power reactors to Finland: safeguards, security and safety considerations in design

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Abstract:

The early interaction between the stakeholders in nuclear non-proliferation and security issues can be initiated like in the nuclear safety assessment. The importance of safeguards and security considerations should be incorporated into the regulatory requirements to facilitate the 3-S concept. The communication between the IAEA, RSAC, SSAC, the operator/ordering company and the designer side shall opened in transparent and mutually acceptable way after the decision to build a new facility and continued in an iterative process during the construction. In summer 2010, Finnish Government made Decision-in-Principles to authorise the construction of two new nuclear power reactors and the expansion of the geological repository for spent nuclear fuel. The planning and designing of these facilities will be regulated according to the new principle to incorporate security and safeguards considerations to the design of new nuclear facilities.
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Keywords: new nuclear reactor; safeguards, security, design

1. Introduction

Nuclear safeguards is commonly acknowledged to be the traditional nuclear material accountancy and reporting system and the main stakeholders are the international, regional and local authorities, and the operators. The introduction of the Additional Protocol (1996) extended the scope of safeguards to the non-proliferation control of nuclear programmes and fuel cycle related activities in Member States around the world. In addition, the United Nations Security Council Resolution 1540 (April 2004) requires every State to ensure that export controls, border controls, material accountancy, physical protection are efficiently taken care of and calling all States to develop appropriate ways to work with and inform industry and the public regarding their obligations. Despite of these demands, it is still common that safeguards culture is acknowledged to cover the stakeholders at national, facility and individual levels in Member States, but not the designers, manufacturers and supply organisations that operate on global market.

The design of any nuclear facility is subject to many economic, technical, legal, security, safety, environmental and other constraints, and it is the function of the design team to find solutions which are optimal within these constraints. Safeguards implementation is an additional factor which should be equally taken into account, preferably during the design stage as indicated by the IAEA already in 1998 [1]. The early implementation of safeguards to new facilities would improve the status of safeguards in the nuclear society.

The planning and implementation of safeguards measures to a new facility begins, in the current approach to safeguards, after the design and construction phase, when the formal Design Information is available to the IAEA. Besides, the IAEA Safeguards Manual updated in 2004 still supports this approach. The time line according to INFCIRC/153 is 180 days before nuclear material is introduced to the facility. Therefore, the operator typically submits this final information with the nuclear licence application. By this time, many, if not all, details of the design are already unalterable. However, the

requirement for early submission of the provisional design information dates back to April 1992[1]. The IAEA executive Board called all the parties to comprehensive safeguards agreements to inform the Agency of their programmes for new nuclear facilities and activities, and for any modifications to existing facilities through the provision of preliminary design information, as soon as the decision to construct, to authorize construction, or modify has been taken. In order to find means to have safeguards involved in the facility design phase, the IAEA initiated in 2008 the safeguards-by-design (SbD) task, but no exact mechanism how to adopt these was specifically addressed in the STR-360 report of the task [2]. The findings are currently evaluated within the support task.

The importance of designing facilities to be secure and proliferation resistant should be understood by all stakeholders. The well functioning security and safeguards systems would reduce the burden to the operator and make the estimated operational costs more reliable. Also, taking the safeguards requirements into account during the design process helps to assure the operator and constructor that there will be no unexpected changes required in the later stages of the construction process due to unexpected safeguards demands. This is potentially a very useful help in project risk management during the construction. However, the attitudes, practices and legislative bureaucracy do no support the effective incorporation of necessary provisions for safeguards during the design and construction phases.

The experiences from the nuclear construction project in Finland [3] indicate that the IAEA safeguards requirements have to be communicated to the designer through the national authorities of a State and finally the company contracting the facility design or construction. The construction of new nuclear reactors was authorised in Finland in summer 2010. The legislative frame work will be amended according to the experience from the current practices and the new projects are seen as new challenge to develop modern safeguards culture between the stakeholders.

2. Safeguards requirements in the licensing procedure

Nuclear safeguards are applied to all materials and activities that can lead to the proliferation of nuclear weapons or sensitive nuclear technology. These safeguards include nuclear materials accountancy, control, security and reporting of nuclear fuel cycle related activities. In order to uphold the Finnish part of the international agreements on nuclear non-proliferation—mainly the Non-Proliferation Treaty (NPT)—this regulatory control is exercised by the Nuclear Materials Section of the Finnish Radiation and Nuclear Safety Authority (STUK).

The national nuclear safeguards derive their mandate and scope from the Finnish Nuclear Energy Act and Decree. These were amended during 2008 as a result of the general constitution-based renewal of the Finnish nuclear legislation system. The operator's obligation to have a nuclear material accountancy system and the right of STUK to oversee the planning of and generation of design information for new facilities was introduced from STUK regulations to the Decree. During the process of licensing new nuclear installations the operator’s competences are reviewed for the Decision-in-Principle, for the nuclear construction license, and for the operating license.

As stipulated by the Act, STUK issues detailed regulations on safety and security (the YVL Guides) that apply to the use of nuclear energy. The operators’ role in the national system of accounting for and control of nuclear materials is defined in YVL Guide 6.9. Currently, all STUK YVL Guides are under renovation. The guides relevant to safeguards shall be merged into one joint new guide. In addition to these self-standing safeguards requirements, the interaction between safety, security and safeguards requirements are incorporated and cross-referenced in the structure of the new YVL guides. The draft version has already been communicated to the stakeholders. The new Guides shall be issued by the end of 2011.

2.1. Experiences from the Olkiluoto 3 reactor

[1] Source – IAEA Board of Governors Report, GOV/2554/Attachment 2, April, 1992

The Finnish Government granted a licence for constructing a new nuclear reactor, Olkiluoto 3, on 17 February 2005. As a part of the licensing process, TVO's plan for arranging the necessary measures for preventing proliferation of nuclear weapons was approved by STUK. The main focus was to confirm the import licensing for nuclear technology for peaceful purposes. In the review process the competences and capabilities to conduct the non-proliferation control was assess.

The operator prepared a draft for the Basic Technical Characteristics (BTC) of the new unit in 2008. This draft was reviewed by STUK, the IAEA, and the Commission twice in 2008. Thus, the inspectorates had the possibility to plan the surveillance and containment measures in advance. Future actions were, during 2009, waiting for the advancement of the construction of the fuel handling buildings. The commissioning of the OL-3 unit is scheduled to take place in 2013. According to the Safeguards Regulation, the BTC is due at least 200 days before the first consignment of nuclear material is to be received.

The experience obtained from this construction site in Olkiluoto clearly points out the need to bring in the safeguards requirements at an early stage of facility design. The early design information is generated, in principle, before the construction, but when planning the installation of containment and surveillance equipment and cabling, some of the desired precautions were missing because these were not communicated during the design of the fuel handling systems, and in particular, before the casting of containment structures. As experienced, the communication between the IAEA, regional (European Commission) and national safeguards authorities, the operator/ordering company and the designer side should be conducted and continued in an iterative process during the construction – much before the official documentation is available for traditional safeguards implementation.

2.2. Authorisation of new reactors

Three power companies and the spent fuel management company submitted their applications during 2008 and 2009 for a Decision-in-Principle to construct new nuclear power plants in Finland. During 2009 STUK reviewed these applications for nuclear safety, security, safeguards etc. as defined in the Nuclear Energy Act. The plan and competences to conduct the non-proliferation control was reviewed by STUK similar to the review for the Olkiluoto 3 case.

The construction of new nuclear power reactors at Olkiluoto and at one of two sites proposed by Fennovoima in northern Finland, were approved by the Government on 6 May 2010 and endorsed by the Finnish Parliament on 1 July 2010. The new applicants are requested to submit a nuclear construction license application within 5 years, and the new stakeholder a plan for its waste management within 6 years time.

The experiences obtained from the regulatory oversight of the current construction projects at Olkiluoto clearly point out the need to bring in the safeguards requirement at an early stage of facility design. Therefore, the safeguards systems for these new facilities shall be communicated and designed in accordance with facility design before the nuclear construction license application. The STUK requirements in defined in the YVL Guides will be amended in such a way that the companies shall prepare the preliminary BTC before the construction license phase to enable the incorporation of the cabling and installation of safeguards instrumentation parallel with the design of other nuclear instrumentation.

3. Conclusions

The introduction of safeguards into the design of new facilities should be incorporated in the early design stage. Safeguards instrumentation is typically mounted at old facilities after the commissioning of the facility. The IAEA safeguards and security requirements are typically not known to the designers, who have to consider, first of all, the nuclear safety requirements. Therefore, the interaction between these stakeholders should be arranged, e.g. via strong national authorities.

The safeguards agreements call for an early submission of preliminary design information in order to facilitate the planning of facility specific safeguards. The integration of containment and surveillance as well as monitoring systems and non-destructive assay equipment to nuclear material transfer system should be included in the facility design. It is obvious that the early introduction of safeguards during the design phase could reduce the impact of safeguards on the cost and facility operations and help to mitigate the risk of surprising, and possibly costly, changes required to fulfil the safeguards requirements.

In reality, submitting binding, official and complete Design Information documents before the design is finalised is not possible – and by that time many of the details that make or break the cost-efficiency of the facility safeguards are already fixed. The early interaction between the stakeholders can be initiated like in the nuclear safety and security community. It is necessary that safeguards and security related requirements will be incorporated into the regulatory nuclear design requirements, like those of nuclear safety. The amendments in the Finnish nuclear legislation combine the safeguards, security and safety requirements for the consolidated oversight of the use of nuclear energy.

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Safeguards By Design – As applied to the Sellafield Product and Residue Store (SPRS)

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ABSTRACT

Sellafield Product and Residue Store (SPRS) is a new facility that has been constructed on the site of Sellafield. The design work started in early 2001 and active commissioning commenced with the introduction of the first nuclear material which arrived in the building early 2011. The store has been designed for the long term storage of Plutonium product (PuO2) from Thorp and Magnox, MOX residue powder from Sellafield MOX Plant (SMP) as well as pellet, powder or granular PuO2 residues from the older stores on the Sellafield site.

This paper describes the application of Safeguards By Design commencing at the early design stage based upon the Safeguards Approach to be applied by DG ENER at the Sellafield Product and Residue Store (SPRS). The approach had been developed based upon the requirements for implementing Commission Regulation 302(2005) and the technical measures to be implemented in order to meet Article 77(a) of the Euratom Treaty. In order to meet these requirements a close dialogue was established between the different interested parties and the design team for the installation of instrumentation with associated cabling in order to implement the agreed safeguards measures. Early contacts at the design stage facilitated the inclusion of installed safeguards supplied instrumentation into the overall design and facility construction. The equipment and cabling supplied by Euratom was incorporated into the planning and construction phases. This ensured that upon plant completion the safeguards tools were commissioned and ready for the verification of the first nuclear material to be introduced into SPRS. Detailed discussions at the early stages of the design phase raised the profile of nuclear material safeguards and made certain that the necessary instrumentation infrastructure was incorporated into the plant infrastructure.

Keywords: Sellafield; plutonium; nuclear safeguards; SPRS (Sellafield Product & Residue Store)

1. Introduction

1.1. General

The Sellafield Product and Residue Store (SPRS) is a new facility that entered into operation earlier this year (2011) for the long term storage of the plutonium product generated from the reprocessing operations on the Sellafield site. The design work commenced in early 2001 and active commissioning commenced in 2011, with the first nuclear material arriving in the building in February 2011. The store is planned to go into full operation later this year after the initial test loading phase. It has been designed for the long term storage of the Pu products from the Sellafield site reprocessing plant as

well as MOX powders from Sellafield MOX Plant (SMP) and a variety of plutonium residues from the older stores on the Sellafield site. The store has been constructed for a capacity for 9,600 cans but its modular design allows for future expansion to increase the storage capacity.

Discussions on safeguarding this plant, commenced at an early stage prior to the start of construction, between the Safeguards Inspectorate, the United Kingdom Safeguards Office, and both the design team and the future facility operator. This ensured that the design team and the future operational team were aware of the nuclear material safeguards requirements and the proposed safeguards measures that would be implemented into the plant by the EURATOM safeguards inspectorate. The inspectors were able to define clearly the expected boundary of the nuclear material balance areas based upon previous experience and the inspection strategy to be implemented. A detailed examination of the proposed route of the passage of nuclear material through the plant allowed the inspectors to indicate the possible locations for the unattended measurement and surveillance systems. This dialogue on the design concept enabled the safeguards authorities to highlight the need for a dedicated unattended measurement station to monitor all the nuclear material items entering and eventually leaving the store. The space and access requirements for the proposed location of this device were thus factored in at an early stage. This measurement feature was one of the main cornerstones of the safeguards requirements. DG ENER presented an overall draft safeguards approach, based upon the current safeguards implementation guidelines, to identify the potential measurement points and the proposed locations for the different monitoring instruments (cameras, seals, neutron monitors, etc). Understanding the route to be taken helped the inspectors to define the usual containment and surveillance features for cameras, seals, and door monitors. The dialogue with the design team enabled DG ENER to settle the boundaries of the proposed Accountancy Areas together with the necessary surveillance hardware.

Another important aspect was the installation of a capability to receive key measurement data, surveillance images, as well as electronic seal status through a secure line to a dedicated receiving station in Luxembourg. The signals and data of the safeguards equipment was designed to be routed to a dedicated data collection room inside the facility where the key data elements could be transferred via a dedicated line to headquarters. This remote data transmission link has enabled the inspectors to follow carefully certain commissioning activities related to the installed safeguards instrumentation as well as allow the preparation of inspection activities.

2. General design and operating features

For safeguards purposes it was agreed that 100% of the cans entering into the store will go through one of the two installed automated Neutron-Gamma measurement stations. This operational constraint was necessary as the product and residue material will be delivered from different parts of the Sellafield site. In order to regain the knowledge of the material as it passes into the SPRS store a measurement of the can contents using one of the two Can Contents Monitors (CCM) will be performed to facilitate this. In conjunction with these measurement stations are can identity readers. These readers are able to individually identify each particular can after they have been presented to one of the measurement stations. The measurement output from the CCMs will be branched so that DG ENER can independently calculate the measurement values for verification purposes.

Cans are stored in channels within the store. Once they arrive in these channels their re-verification becomes difficult in practical terms due to the large number of can withdrawals and can reshuffling that would be necessary in order to access specific cans. Therefore multiple Containment and Surveillance (C/S) will be applied to retain the continuity of knowledge of cans and thereby reduce the requirements for subsequent re-verification. There will be a very limited number of re-measurements during the annual Physical Inventory Verification (PIV) as part of the overall assurance scheme.

Sub-perimeters have been set up within the main C/S perimeter to allow greater operational flexibility and to reduce the effects of major C/S failures. These arrangements would allow man-entry to a part of the C/S zone whilst preventing access to the main store. The C/S system is automated as far as is possible to reduce inspection effort.

The proposed lifetime of the plant extending to possibly 2120 means that most of the nuclear material inventory will remain in the building in a static state after the initial loading. As such the overall approach to the inspection regime will need to be re-examined after the store has been filled to reflect this static state.

A particular issue with the SPRS plant is the passive cooling of the storage channels. In order to prevent the possible movement of material within the interconnecting air passages upstream and downstream a number of actions have been taken during construction. These include the installation of a number of physical barriers, preventing access via the plenum or air inlets and the use of appropriate technical means to provide assurance that these barriers have not been removed. In addition containment inspections may be carried out on a short notice basis to confirm that the necessary measures undertaken during the construction phase have not been modified or altered.

3. Design verification

3.1. General Scheme

To achieve the Safeguards Objectives, the Safeguards Approach is based upon an initial verification during the construction phase and subsequent re-verification of the Basic Technical Characteristics (BTC) during the annual Physical Inventory Verifications including the use of the 3D laser scanner [1] to check the absence of changes in previously identified key locations.

3.2. Initial verification

The initial verification of the BTC took place as the construction was completed and prior to closure of each particular location. For the ducting and ventilation photographs were taken and stored on site for future reference. Upon completion of the design verification for each particular location seals were applied to ensure that the knowledge was maintained.

The use of the scanner for design re-verification will form part of the annual PIV design verification activities. This activity formed part of the initial BTC verification at the zero PIV prior to the introduction of material at the beginning of 2011.

3.3. Ventilation Ducting

There is a need for surveillance of the ventilation ducts due to their size and the possibility of a diversion scenario for removal of cans from the inside of the store. In order to accommodate this aspect certain controls were chosen including the use of a 3D laser scanner device, developed by the JRC Ispra, on the grille of the outlet ventilation duct, to be able to detect any form changes over time. A reference set of scans were carried out during 2010 and a random selection were re-verified during the beginning of 2011. The results indicated that there had been no changes over this short period. All the data is kept in a secure location within the building itself and is available to the inspectors on the occasion of the PIV and other BTC verification inspections as required.

3.4. Store

Due to the complex nature of the internal part of the store where the storage channels are located the use of a 3D scan to provide a reference 3D picture was made. This scan allowed the inspectors to confirm the integrity of all the openings on the walls and ceilings inside the store associated with the ventilation and support services on an ongoing basis. It is intended that subsequent 3D laser scan usage and follow-up will form part of the BTC verification performed at the annual PIV.

3.5. Safeguards Scheme

The safeguards scheme is based upon two accountancy areas with one covering the transit, handling, and inspection area for all nuclear material movements into the building and the second one the store itself. All nuclear material movements into the store will pass through one of the two installed CCMs supported by a combination of cameras, seals, identification readers and monitors providing a sound surveillance boundary.

4. Inspection Activities

4.1. Introduction

The inspection approach is based on the legal requirements of Art 77a of the Euratom Treaty and Commission Regulation *302(2005).* The inspections will be carried out in accordance with the implementation paper of the Commission reflecting the provisions foreseen in the guidance paper entitled 'A new framework for Euratom Safeguards' discussed at the Working Party on Atomic Questions of the Council of the European Union in December 2005 [2]. These inspection activities would be as follows:

- An annual Physical Inventory Verification (PIV).
- BTC verification during the PIV, including any declared modifications.
- A check of the Nuclear Materials Accountancy & Control (NMAC) records during the PIV.
- 6 – 11 interim inspections

4.2. Physical Inventory Verification (PIV)

Use will be made of the possibilities for in situ verification of cans to provide assurance that cans are present as declared in the channels.

The operator's physical inventory listing will be verified once per calendar year at intervals of not more than 14 months. The following activities will take place:

- Verification of the list of inventoried items (LII).
- For the material which has been under C/S up to 5 items may be verified for gross and partial defects
- The use of the installed Cd-Te detector on the transfer trolley to verify in situ a number of randomly selected cans that are present as declared in the channels.
- Examination of accounting and operating records, and supporting documents for correctness and self-consistency.
- Establishment of updated book inventory.
- Verification of receipts and shipments.
- Verification of the BTC.
- Use of the 3D laser scan to verify absence of any changes or modifications to the specific plant locations.
- A review of C/S measures.
- Servicing of surveillance devices if appropriate.

4.3. Interim Verification activities

A number of interim inspections will be carried out between the annual PIV inspections. These will number between 6 – 11 inspections,

Interim verification inspections will be an important feature during the early stages of the plant life as there are expected to be many movements into the store. These inspections will be used to verify the flow of material and build on the knowledge from the annual PIV. The activities will include the verification of the nuclear material contents based upon the measurements through the can contents monitor and the related surveillance recordings.

5. Instrumentation

5.1. Can Contents Monitor

The Can Contents Monitor has been designed into the plant so that the contents of the Pu cans being introduced into the store can be verified prior to transfer into the dedicated store location. Discussions at the early design phase ensured that there was sufficient space in the transfer route for both the measurement station and the can identification system. These items are located in a dedicated room that is sealed and equipped with video surveillance to serve as a back–up in the case of equipment failure. There are transfer track openings that are monitored to confirm the direction and movement of the cans. The Can Contents Monitor is based upon a passive neutron coincidence counter (PNCC) with multiplicity analysis in order to determine the amount of spontaneous fission isotopes (mainly 240Pu and the other even isotopes of plutonium) present in each can. It uses High Resolution Gamma Spectrometry (HRGS) in order to determine the isotopic composition of the plutonium in each can. Furthermore it has been designed to allow analysis of all different cans and material types which are expected to be stored in SPRS and is thus quite versatile.

There are two identical monitors installed in SPRS on the two different floor levels to handle all the material movements into the store and any occasional transfers out.

Within the inspection scheme, the monitors will be used to verify the flow of material into and out of the store material balance area. All cans which enter and leave the stores will be quantitatively analysed. Furthermore the CCMs will be used during the physical inventory verification to re-measure a small number of selected cans randomly selected as required. The units can also be used to provide a potential containment backup solution in the case that the containment and surveillance scheme of the stores should ever be compromised

The data acquired at the monitor stations will be collected with the Commission developed data acquisition system (Remote Acquisition of Data And Review system). The data will include the neutron coincidence or multiplicity measurements, the gamma spectra and details of the can identification. The analysis of the data will be performed using a specific evaluation package with the acronym CRISP (Central Radar Inspection Support Package), a Commission (DG ENER-E) developed data evaluation package. The CRISP software correlates data of different sensors, calculates the measurement results and material flow paths and compares these with the operator declarations. CRISP finally provides a report for the inspector. [3]

5.2. Surveillance scheme – video and neutron monitors

The store part of the facility will be covered by a multiple containment and surveillance system. This will employ a combination of neutron monitors and surveillance cameras that will allow the inspectors to be able to follow the flow of nuclear material into, out and through the store The final details of application of containment and surveillance could only effectively be applied upon completion. The containment and surveillance scheme took advantage of the containment provided by the physical barriers of the plant and the inspectors ensured that they were satisfied with the integrity during and upon completion of the construction phases. Additional confidence was assured with the application of seals and use of the 3D laser scanning at the final commissioning stage.

The camera views were fine tuned when all the construction and commissioning scaffolding and other obstructions had been removed. The use of computer simulation models helped to identify the locations for the installation of cameras but the final confirmation still needed to be made during the final commissioning stages. Sealing locations were readily identified and preparations made for wire or cable application well in advance.

Knowledge of the Pu can arrivals into SPRS will commence when they reach the CCM and are subjected to measurement. Their subsequent movement will be monitored and recorded both by digital surveillance cameras and 17 neutron detectors mounted at strategic points inside the storage halls. The high sensitivity neutron monitors [4] to be employed will be similar to those already successfully used in a number of Pu handling facilities within the European Union. Their sensitivity means that they are readily able to recognize movement of items containing nuclear material. The recorded signals can be analysed automatically using a data analysis system and give the inspector a full interpretation regarding the path of movement, which can be compared with the declaration.

As well as the neutron measurement system and neutron monitors an independent video surveillance system (FAST system), consisting of 34 digital cameras has been installed. The individual cameras have been sited to cover known nuclear material movements as well as possible diversion routes out of the secure area. The cameras help to provide the containment layer coupled with other monitoring devices such as seals and door closure monitors.

5.3. Electronic Seals

In order to reduce the number of inspection activities and enhance the containment of the different possible access routes seals will be applied as much as possible. The inspector presence in SPRS will probably total only a maximum of 40 days per year so it is important that seals are effectively applied to help maintain knowledge of the store between visits.

One important area of application of electronic seals will be on the emergency exits, which, in principle, should be rarely used. Under normal operating conditions it is not anticipated that the channels will be sealed, nevertheless preparations have been made. The seal of choice for these applications will be the EOSS seal [5]. This seal can be remotely read out and has been designed with a high level of reliability and security. The readout of the seals will be via the RADAR system and the analysis of the collected data by CRISP. Cabling for the interrogation of the EOSS seals was installed during the construction phase including the capability for interrogation from headquarters.

5.4. Laser Verification

During the discussions at the design and construction phases of SPRS it became clear that due to the size of the ventilation ducting and possible diversion scenarios for removal of cans it was important that the containment of the store needed to be checked for its completeness. One particular area that required additional attention was the store cooling ducts that were routed from the 0m level walkways up to the roof level and exhausted into the atmosphere through a series of ventilation stacks. In order to restrict possible access into these stacks the duct outlets were each sealed with a metal grid structure. To ensure that there has been no tampering of this structure the integrity of the upper security grille at the base of each stack required checking. A Laser Verification technique, developed by JRC Ispra [1] for design re-verification has been applied. This technique based on a 3D scan of the structure will check the layout of the ventilation stack outlet and will confirm whether these grids have been altered or tampered in anyway since the reference scan.

5.5. In situ Channel monitor

The cans are placed in storage tubes in the main store on either the 0 m or the 6.35 m level by one of 4 automated charge machines. There is one charge machine for each charge corridor with 4 corridors making up the present SPRS store configuration. In order to provide the inspectors with the assurance that the cans are present as declared in the channels an in-situ verification method has been implemented in the safeguards approach. A number of channels will be selected randomly at the annual PIV to confirm the number and presence of cans as declared within the selected channels.

The in-situ inspection verification of the cans stored within the channels will be by the introduction of a Cd-Te detector that will pass under the row of cans and confirm the presence of nuclear material. The signal from the detector will be interpreted using a standard MiniMCA gamma spectrometer or its new digital upgrade [6]. There will be an in-situ inspection monitor installed on each one of the 4 charge machines.

5.6. Can Identification Verification

When the plutonium cans are introduced into the store they will be identified and tracked using a combination of an Optical Barcode(OBCR) and/or an Optical Character Recognition (OCR) System as they pass along the transfer track after the CCM. The OBCR system will read either the barcode or the alphanumeric identifications on the cans as they are transferred into and, on the rare occasion, out of SPRS. The system will be a combination of cameras reading the numerical characters on the outer Pu cans and the logic image processing algorithms. The systems installed at the moment rely upon video image recognition for both the barcode as well as the alphanumeric characters. In the long term it is proposed to replace these systems with a laser reader device that will avoid the need for possible human intervention in interpreting the visual images.

The original intention was to branch the signals of the operator for the can identities but due to a possible conflict in using the plant data network independence had to be preserved. The final decision on this approach came late in the construction and commissioning schedule which necessitated the duplication of the can identification systems. After the installation of the OBCR and OCR cameras and following a series of discussions with JRC Ispra on how to handle and interpret the visual images from these cameras it was decided in the medium term to replace all the cameras with a laser reading system that would be more robust and reliable.

6. Data Transmission

The Regulation (Euratom) N° 302/2005 advocates the use of information technology and of telecommunications networks in the exchange of data between the Commission and operators. The changes to the on-site verification frequency to plants within the European Union since 2005 has re-enforced the need for optimizing the inspectors' work during on-site inspections and the transfer of part of the verification activities back to headquarters in Luxembourg. The development of this idea implies the need to transfer elements of data to headquarters that were previously only accessible in the installations. Well organized operating records, well structured operators' databases and well defined transfer formats of these data to the DG ENER inspectors are key issues for an efficient use of such data in Luxembourg.

The installed and integrated instrumentation and equipment form a key aspect of the safeguards arrangements within SPRS. The data transmitted off site back to Luxembourg is structured and targeted so that the best use can be made of this information in both the preparation for an inspection and the subsequent inspection and evaluation process. The secure link which is used between Sellafield and Luxembourg has been described earlier [7]

During the commissioning phase it has been extremely useful in having certain key data available in Luxembourg to help commission the different instruments as well as assist in the calibration of certain key items.

The on-site inspections activities are optimised by analysing the transmitted signals of the electronic monitoring, electronic seal status, selected CCTV images signals and the CCM measurement system as well as some other key monitoring devices such as door monitors from the plant back to headquarters.

The use of inspection data transmitted back to headquarters enables DG ENER to modify the modalities of the inspection verification scheme as the store becomes full and nuclear material transfers into and out of the store become infrequent. This anticipated improvement in efficiency may result in either reduced number of inspection man days or the number of visits per annum as well as increasing the flexibility for access to the store during routine operations.

7. Conclusions

The early interaction between the designers and the safeguards inspectors involved in the SPRS project has helped as follows;

- The safeguards approach was defined sufficiently early so that the instruments could be built into and incorporated into the design

- The instrumentation was installed during construction to ensure signal independence.

- Design information verification was successfully carried out during the different construction and commissioning phases.

- The involvement from a very early stage ensured that the DG ENER inspectors were able to appreciate and understand the proposed plant layout and plant operations. Using this knowledge the inspectors were able to identify possible diversion routes.

- The involvement of the inspectors at an early stage in the design process heightened the awareness of the plant designers to the safeguards requirements, techniques, and instrumentation.

This allowed them to integrate the proposed safeguards equipment in an optimal and cost effective way.

- The close co-operation between the plant designers and the safeguards authorities, during the detailed design phases, ensured that the inspectors benefited by having their instrumentation designed into the plant in the most advantageous manner.

- Incorporation of Remote Data Transmission into the design and the necessary appropriate measures for the implementation of security of the safeguards information.

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12 Containment and Surveillance

Non-Traditional Surveillance Systems and their Application to Safeguards

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Abstract:

The term "surveillance" is most often associated with images or videos captured in the visible region of the electromagnetic spectrum – approximately 400 to 750 nanometers, as this is where the human eye can see. However, the electromagnetic spectrum is vast and possibly other information can be extracted using systems that are sensitive to other wavelengths or techniques that could exploit what has been non-traditional information. This additional information might be useful in applications where lighting is low or obscured; for heat detection or analysis; to detect particular chemicals, effluents, or materials; to detect objects through-the-wall; to determine polarimetric behaviors of light with materials; or for general improvement in discriminating an object of interest. For instance, thermal cameras can detect emitted heat, which may allow for better material or object discrimination in an image, or even for determining density variations in materials. Other imaging techniques, even in the visible region, can be exploited to process information in atypical ways. This paper will look at commercial or academic, non-traditional imaging systems and evaluate possible safeguards applications.

Keywords: surveillance; non-visible imaging; electromagnetic spectrum

1. Introduction

Containment and Surveillance (C&S) methods in safeguards contribute to "continuity of knowledge" regarding nuclear materials and inspection equipment. Surveillance has traditionally relied upon imaging systems sensitive to the visible region of the electromagnetic spectrum. However, imaging systems are available which exploit other regions of the electromagnetic spectrum as well as other physical quantities associated with an optical field, and exploring these other regions and quantities has the potential to strengthen safeguards activities by offering additional information for improved surveillance measures.

The Next Generation Safeguards Initiative [1], or NGSI, is meant to strengthen international safeguards, coordinate the U.S. safeguards technology programs, and revitalize the U.S. safeguards technology and human capital base. NGSI has five pillars: (1) policy development and outreach, (2) concepts and approaches, (3) technology development, (4) human resources development, and (5) international infrastructure development.

The goal of the 3rd pillar, "technology development", is to enable the IAEA to optimize the use of limited human and financial resources. Its three objectives are to develop advanced tools and methods to detect diversion of declared nuclear materials, develop advanced tools and methods to detect undeclared production or processing of nuclear materials, and provide information analysis solutions to improve state level assessments. It is within this technology development pillar that non-traditional imaging could be added as a new tool set.

From the European perspective, specific tasks from the ESARDA C&S Compendium [2] that warrant exploration of non-traditional imaging are task (1) advise the European Commission and IAEA on new and improved instruments and methods and on areas where R&D effort is still needed, and task (10)

study technical characteristics of instruments and devices from other domains (e.g., physical protection) and investigate the possible transfer of technology from these domains to the safeguards area.

So what is non-traditional imaging? The answer crosses multiple physical domains – spatial, spectral band, spectral resolution, time, and direction, to name a few, and thus this introduction will begin with a brief explanation of the limitations of visible imaging.

The visible region of the electromagnetic (EM) spectrum (Figure 1) ranges approximately from 400 to 750 nm. What humans perceive as color is the wavelength dependence of the radiation reflected from an object within this region. Under bright illumination, the human eye has three classes of cone photoreceptors – one for long visible wavelengths (red), one for medium visible wavelengths (green), and one for short visible wavelengths (blue). The limited variations of color that humans see are combinations from those three cones. Conventional visible region digital cameras[1] measure the intensity of optical radiation reflected from a scene on a detector. The source of optical radiation often is broad spectrum, meaning that it contains radiation outside of the visible spectrum. Many detector materials within digital cameras are sensitive to the radiation outside of the visible band as shown in Figure 2; however, filters are often placed on the camera to eliminate this radiation as it causes unwanted effects in the imagery. To image just outside the visible region of the EM spectrum, the filters used on digital cameras are removed. Further away from the visible region, new detector materials and technologies are required – as well as new illumination sources for some EM bands.

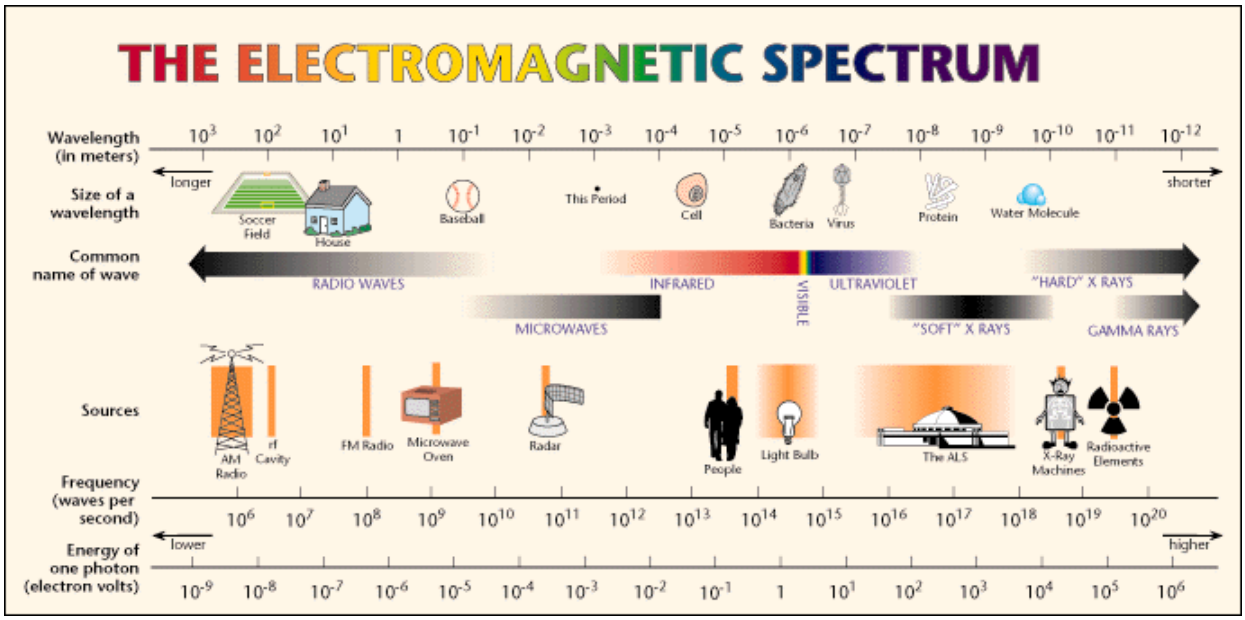

Figure 1: The electromagnetic spectrum (Courtesy: Remote Sensing Tutorial [3]).

[1] Common visible region digital cameras as systems will simply be referred to as digital cameras, although other types of imaging systems use digital technology as well. Detectors for each type of imaging system will be specified in more detail.

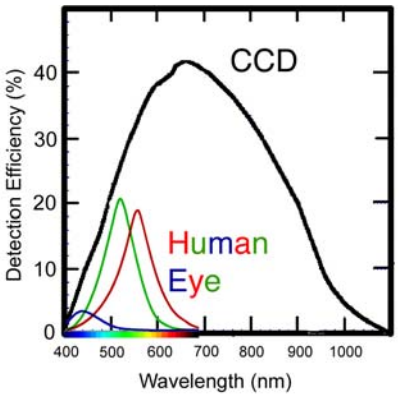

Figure 2: Human eye detection efficiency versus Charge Coupled Devices (CCD) commonly found in digital cameras [4].

2. Imaging Systems across the Electromagnetic Spectrum

2.1. Reflective Infrared

Cameras that provide images in the near infrared (NIR) are sensitive to light just beyond visible red light – wavelengths in the range of 750 to 1000 nm[2]. NIR camera images require some means of illumination either from broad spectrum natural sources such as sunlight, moonlight, night sky light or from active NIR illuminators.

Two detector technologies are most commonly employed to capture images in the NIR – Charge Coupled Devices (CCDs) used in digital cameras and Indium Gallium Arsenide (InGaAs). CCD cameras are sensitive to light in the 350 to 1000 nm range, which overlaps the ultraviolet, visible, and NIR bands. InGaAs cameras are sensitive to radiation in the 900 to 2500 nm (0.9 to 2.5 micron) ranges, but are more common to approximately 1700 nm (1.7 microns).

The performance of a CCD camera in NIR applications is affected by the quantum efficiency (QE) or sensitivity of the imager at NIR wavelengths. Imager relative QE's can range from 0 to 85% in NIR wavelengths as compared with on the order of 100% in the visible spectrum. When selecting a CCD camera for NIR imaging, choosing a camera with high NIR QE provides the best NIR images at the lowest NIR illumination. The QE of CCD cameras tends to be high at the low (visible) end of the NIR spectrum and significantly decreased at the high end of the NIR spectrum. InGaAs cameras exhibit low QE at the low end of the NIR spectrum and higher efficiency at the high end of the spectrum. Figure 3 shows the QE curves of the silicon detector used in CCD cameras versus the InGaAs detector.

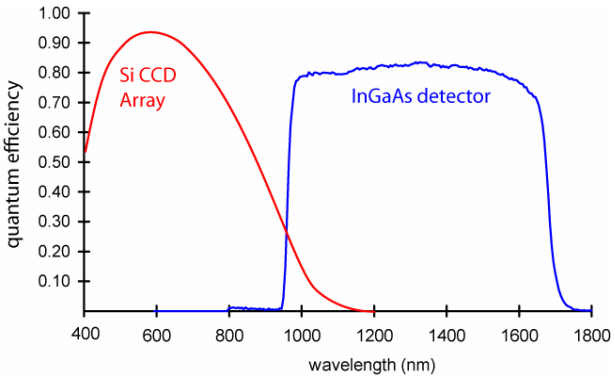

Figure 3: Quantum efficiency (sensitivity) of a silicon detector (CCD) versus InGaAs detector [5].

[2] The numeric values given to regions of the electromagnetic spectrum often depend on the particular application and are both approximate and varied.

NIR sensitive CCD cameras with NIR illuminators are commonly used in physical security applications where discreet observation of a location is required. In interior locations, low power NIR illuminators can provide sufficient illumination to obtain an image similar to that obtained with normal visible room lighting. For safeguards, CCD cameras with NIR illuminators might prove useful in low light applications rather than discreet observation.

Another technology that commonly uses the NIR band is multispectral imaging – particularly for adding NIR capabilities to visible color imagery. One such camera system [6] uses a beam splitter prism to separate light into 4 four CCD sensors, one each for red, green, and blue, and one for the NIR band. The camera is a line scan camera, with 2048 pixels per line requiring image formation by scanning or motion. Applications include fruit and vegetable sorting, food inspection, tiles inspection, print inspection, and PCB inspection. For safeguards, a multispectral line scanner might be useful for item inspection or tamper-indication; a multispectral area camera could be useful for surveillance in low light applications or other applications involving broad differences between visible and NIR (for example, moisture content or seeing through printed material [7]).

The short-wave infrared (SWIR) band ranges from approximately 1000 to 2500 nm. InGaAs cameras can be used above 1.7 microns by varying the fraction of indium in the ternary compound. However, more common detector technologies are HgCdTe (also called MCT) and InSb.

Some applications in the SWIR include low light level imaging, thermal imaging of hot objects (in the 200C to 800C range), vision enhancement, and process control. SWIR is often used for spectral characterization in imaging spectroscopy, which will be discussed later in this paper. These applications may apply to safeguards as well.

2.2. Ultraviolet

The ultraviolet (UV) band extends from 10 to 400 nm, although only the region from 200 to 400 nm is used for imaging because air is opaque below 200 nm. The region from 300 to 400 nm is called the near-UV, and from about 250 to 280 nm the deep-UV. The most common UV illumination sources used for imaging are direct sunlight, gas discharge (black light) lamps, UV LEDs and electronic flash. UV imaging has experienced increased application in recent times due to advances in UV camera hardware technology.

Many CCD cameras used for visible imaging can actually image in the near-UV; however, this is an undesirable feature in visible imagery as the camera cannot simultaneously focus both bands of light and the resulting image would have artifacts such as purple halos around point sources and overall image softening [8]. Because of these effects, CCD cameras employ UV blocking filters. To image in the near-UV, the blocking filters must be removed. The near-UV band can also be transmitted through standard glass lenses; however, the less expensive glass without antireflection coatings has better performance in the near-UV.

Special detectors and lenses are required to image in the deep-UV. The detectors can either use very thin silicon substrates or wave-shifting coatings. Lenses need to be made from quartz (fused silica) or calcium fluorite [8].

There are two approaches for obtaining UV images: reflected UV and UV fluorescence. In reflected UV imaging, the surface of an object is directly illuminated using a UV illumination source. The UV light striking the object's surface is then reflected or scattered (or even absorbed) and detected by a UV camera. In UV fluorescence imaging, the surface of an object is also illuminated using a UV illumination source. However, the object absorbs the UV excitation and reradiates the light at a longer wavelength for detection by visible or IR cameras.

In reflected UV imaging, features and characteristics of an object can be observed that are not distinguishable using other illumination and camera technologies. UV light is scattered more readily by material surface features than light at longer wavelengths (visible, infrared, and beyond). As shown in Figure 4, a slightly scratched plastic surface will appear smooth when viewed in visible light, but under UV illumination, the scratches become pronounced.

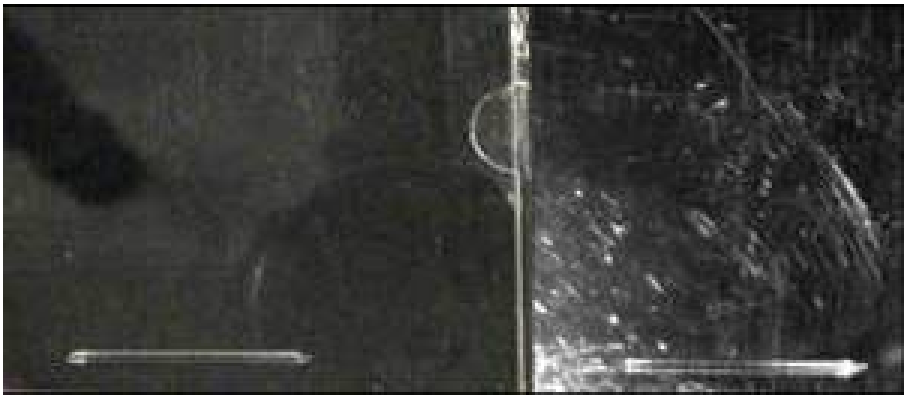

Figure 4: Images of CD jewel case under visible (left) and 365 nm UV light (right) [8].

In a different manner, many materials readily absorb UV light, allowing detection of surface contamination. Furthermore, new and old paints can be distinguished in the UV while appearing identical in the visible spectrum. Figure 5 shows a vehicle that has had its front fender replaced in both visible and near-UV images. The difference in the age of the paint is clear in the UV-image.

Figure 5: The top picture shows a vehicle whose front fender has been replaced imaged in the visible band, and the bottom picture shows the same vehicle imaged in the 320 – 400 nm UV band [8].

UV fluorescence is commonly used in anti-counterfeiting initiatives. Security elements only visible under UV illumination, not in the visible, are incorporated into objects or documents to validate their authenticity. For example, U.S. currency uses UV security threads in its $100 bill [9] which can be verified using a black light, or other UV illumination source. The U.S. state of California has implemented UV photos that fluoresce in the visible band under UV light in their new driver's licenses [10].

For safeguards applications, UV imaging might be useful for tamper indication and equipment authentication. This might be passive or active – scratches can be detected on enclosures, or even organic materials deposited from human contact. Active measures may include embed fluorescent materials into equipment and use UV cameras to detect tampering – perhaps a new approach of more interactive surveilled containment.

2.3. Thermal

The thermal band of the EM spectrum technically ranges from 2.5 to 7 microns for the mid-wave infrared (MWIR) and 7 to 15 microns for the long-wave infrared (LWIR); however, only the regions from 3.3 to 5 microns for MWIR and 8 to 14 microns for LWIR are usable for imaging due to lack of atmospheric transmission in the other regions. Thermal imaging cameras are sensitive to the temperature differences between objects in these regions due to emission of thermal energy.

Thermal imaging has advanced in recent years from a high-cost technology only available to military customers, to ubiquitous detectors available in commercial sectors. In the past, infrared detectors had to be cooled to liquid nitrogen temperatures (77K) to reduce detector noise. New detector materials and other advancements in integrated circuits have allowed for uncooled detectors. Microbolometers are one such type of uncooled detector, and have allowed for small, lightweight, and low power detectors. Uncooled detectors also allowed for large capacity production, and the first commercial applications were for night-time driving aids in BMW vehicles [11].

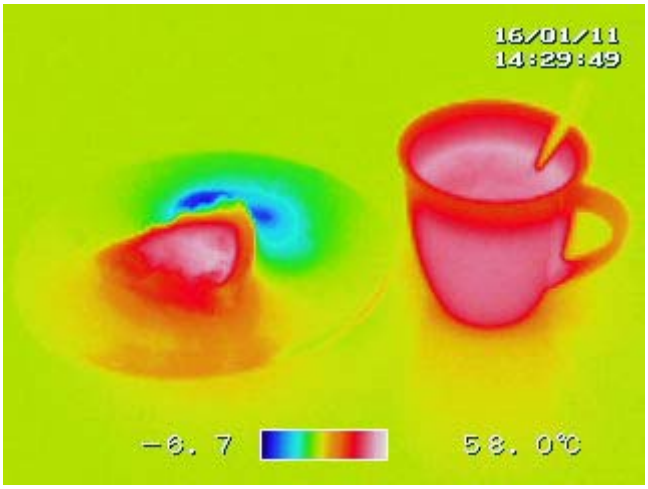

Figure 6: Coffee and apple pie image in the LWIR, using an uncooled detector (microbolometer) commercial thermal imager (Photo courtesy of Pieter Kuiper using an NEC Thermo Shot camera).

Because of the drop in price, thermal imaging is becoming a more affordable option for security and surveillance applications. Standard cameras may rely on auxiliary lighting for illumination, whether from sunlight or active NIR illuminators. Thermal imagers do not require additional illumination as they image differences in temperature from the thermal radiation emission of objects in a scene. This allows for night-time imaging as well as imaging in other poor lighting conditions.

The U.S. Nuclear Regulatory Commission (NRC) has recently required nuclear facilities to provide continuous 24-hour surveillance, observation, and monitoring of their perimeter and control area [12], and thermal imaging is being installed at many facilities for this purpose [11].

Other applications [13] for thermal imaging (including possibly for safeguards) include process control (to monitor quality and safety of plant equipment); analysis of electronics and electrical systems, especially due to overheating or other problems; and analysis of machinery due to heat-producing friction.

Thermal imaging can be used to detect density variations in materials (surface or subsurface non-uniformities or defects to phrase it another way), since continuum heat transfer is impeded by the defects or otherwise affected by density variations [14,15]. Denser materials retain heat longer than less dense materials. Heat can be applied to a scene either through solar loading, or via active means such as brief but intense pulses of light. Infrared cameras are then used to record the surface temperature distribution over time.

For safeguards applications, flash thermography may support safeguards inspections [15] to determine that equipment and facilities are free from tampering and facilities have not been altered.

2.4. Microwave and Millimeter Wave

The microwave and millimeter wavebands have long wavelengths and low energy. The long wavelengths are able to penetrate some materials that visible light cannot since the wavelengths are much longer than the wavelengths of the materials.

The millimeter waveband is defined between 1 and 10 mm. Humans naturally emit enough energy at 3 mm to be detectable, and this energy is able to penetrate clothing. As humans emit this energy through their clothing, any dense objects blocking the energy between the body and a detector will be apparent. Although likely limited for safeguards applications, we mention this as the source of energy is within the long wave bands but still passive.

The rest of this band will require active sources to provide enough energy for detection above terrestrial noise levels. Active microwave sources may generate additional scrutiny by facility operators due to concerns about interference with operator equipment.

The microwave band is from 10 mm to 1 m. Radar technology is within this band, and is well-known for its day/night and all-weather operating conditions. A simple radar system sends out pulses of energy at predetermined intervals. The pulses reflect off of a target, and a portion of the energy is returned to the radar receiver. Geometric and reflecting properties of the target can be estimated from parameters within the returned signal, such as amplitude and frequency. For nonconductive materials, such as vegetation or dry soil, EM waves with long wavelengths are relatively penetrating since their wavelengths are much longer than the material within which they are being transmitted. Ground penetrating radar (GPR) is based on this principle.

There has been discussion about using GPR for Design Information Verification (DIV) in safeguards and thus we provide more detail on this capability [16]. GPR transmits very short pulses into the material, whereupon reflections at dissimilar material boundaries (e.g., steel bars embedded in concrete) return to a collecting/processing system. Generally speaking, GPR operates in the frequency range from 25 MHz (12 m wavelength which falls into radiowaves) to 3 GHz (100 mm wavelength which is in the microwave band). The particular frequency of operation is chosen as a compromise between soil or material penetration depth, object or feature resolution, availability of interference-free frequencies, and antenna and equipment size.

Equipment portability is a key concern when using GPR. Most GPR systems use separate antenna and processing/display systems. This supports multiple frequency ranges, which require different antenna systems, using the same processing unit. For example, a 1600 MHz antenna system provides good resolution of the reinforcing steel in a concrete slab, but a 400 MHz antenna system allows much greater depth of penetration into the material when looking for potential voids below a concrete slab. A separate antenna can also be placed on the material surface, which is needed to couple the maximum amount of energy into and out of the material. Higher frequency systems, i.e., above 800 MHz, are more portable and can be incorporated into a handheld unit. However, these handheld units have a depth of penetration typically less than ½-meter in concrete.

A critical aspect in the practical use of GPR is in the data interpretation. Processed results from current systems are often not intuitive to the novice user, and thus take some training in order to analyze effectively. The processing of a captured time-series of data creates an image on a display that depicts the various layers and objects in the material, subject to the depth and resolution ranges of the frequency and power settings. Figure 7 shows a GPR image of pipes beneath a roadway.

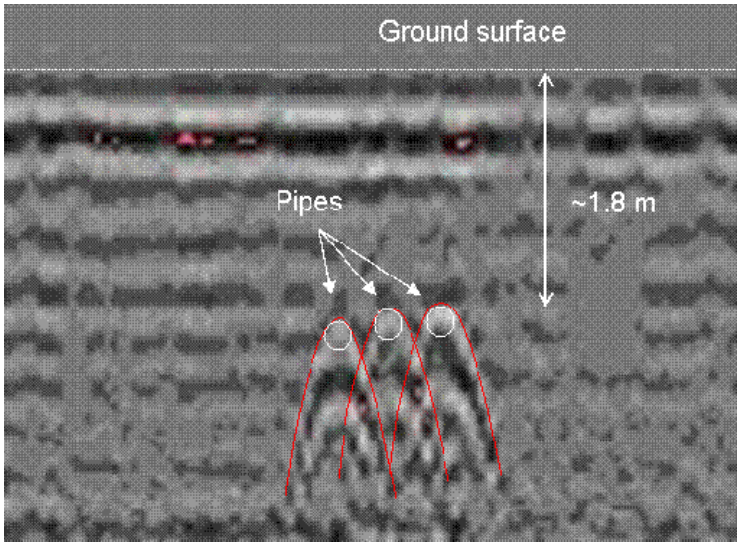

Figure 7: Ground Penetrating Radar (GPR) image of pipes underneath roadway [17].

2.5. X-ray and Gamma rays

X-rays (0.01 – 10 nm) and gamma rays (less than 0.01 nm) are high frequency and high energy, and thus have very short wavelengths. It becomes difficult to measure wavelengths in this region, and thus x-rays and gamma rays are usually referred to by the energy of photons – particles of light that have a particular energy [14].

X-rays and gamma rays can penetrate all materials, and can transfer a significant amount of energy to a localized area producing permanent chemical changes. We saw in section 2.4 that long wavelengths can also penetrate many materials since the wavelength is not on the same order as the material wavelength – however, these long wavelengths cannot image through conductive materials such as water or metals.

X-ray imaging can be used for contraband and anomaly detection – both for imaging through walls and some containers in search of objects as well as human cargo. However, x-ray imaging is typically regulated for human exposure.

Gamma rays occupy the highest energy waveband of the EM spectrum and have highly penetrating properties. They can image where x-rays cannot, such as through steel and other dense materials [14]. Gamma rays are produced by nuclear processes such as radioactive decay.

One method for imaging items inside a steel container [14] is to pass a titanium pellet inside a lead container. The gamma rays are emitted from the lead container through the steel container, and onto x-ray film inside of a light-tight container. Objects from inside the steel container cast shadows onto the x-ray film.

Finally, gamma-ray detectors sensitive to the energy of gamma-ray photons of interest can be used for identification purposes since radioactive materials that emit gamma rays do so at discrete energies. This technique is used in safeguards for identification of different isotopes of radioactive materials for contamination and container contents verification.

3. Acoustic Imaging

Although sound waves[3] are not part of the EM spectrum, we include acoustic imaging due to its ability to image underwater. Under ideal conditions it is difficult to see an object more than 50 m away underwater with visible light [14]. Any deviation from ideal conditions results in drastically worse resolution. High resolution sonar imaging sends an active pulse of sound using a transducer that will also act as the sound receiver. The geometry of the target modifies the shape of the return pulse, while the density of the target affects the strength of the return pulse. The time it takes the pulse to reach the target and return to the transducer determines the distance. The image resolution will depend on the wavelength of the acoustic pulse. One example [14] is a sonar system that uses acoustic pulses with wavelengths of a few millimeters that can resolve features on the order of 20 cm at an imaging range of 100 m.

A search of imaging sonar revealed commercial companies [18] with technologies able to provide high resolution imagery up to 2250 kHz. Figure 8 shows an 11" garden block underwater. The camera is forward-looking, with a slight downward angle.

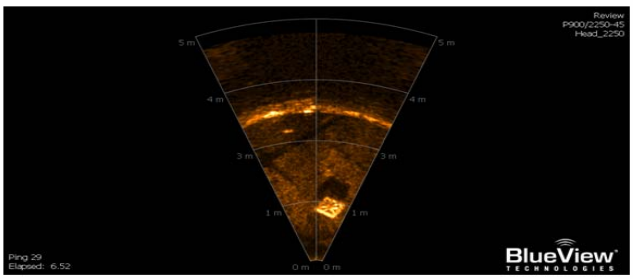

Figure 8: 2D sonar imaging using 2250 kHz sound pulses [18].

Any use of imaging sonar cameras in radiation environments, perhaps such as spent fuel ponds in safeguards applications, would require proper radiation tolerant camera housings. There are many radiation tolerant cameras, housings, and accessories such as lighting available in the commercial sector, including those required for underwater applications.

4. Imaging Spectroscopy

To this point, we have surveyed regions of the EM spectrum, but have not discussed the spectral resolution within these regions. The process by which photons interact with materials (via reflection, scattering, absorption, transmission, and/or emission) varies with wavelength. With enough spectral resolution of a sensor, objects can be characterized by a "spectral signature" – that is the dependence of photons returned to the sensor on wavelength. Figure 9 demonstrates the concept of imaging spectroscopy, or hyperspectral imaging, from a NASA airborne sensor named AVIRIS. A good tutorial on imaging spectroscopy can be found at [19].

[3] Light is composed of transverse waves in an electromagnetic field and requires no medium to travel. Sound is composed of longitudinal waves and requires a sufficient medium (solid, liquid, or gas) to travel.

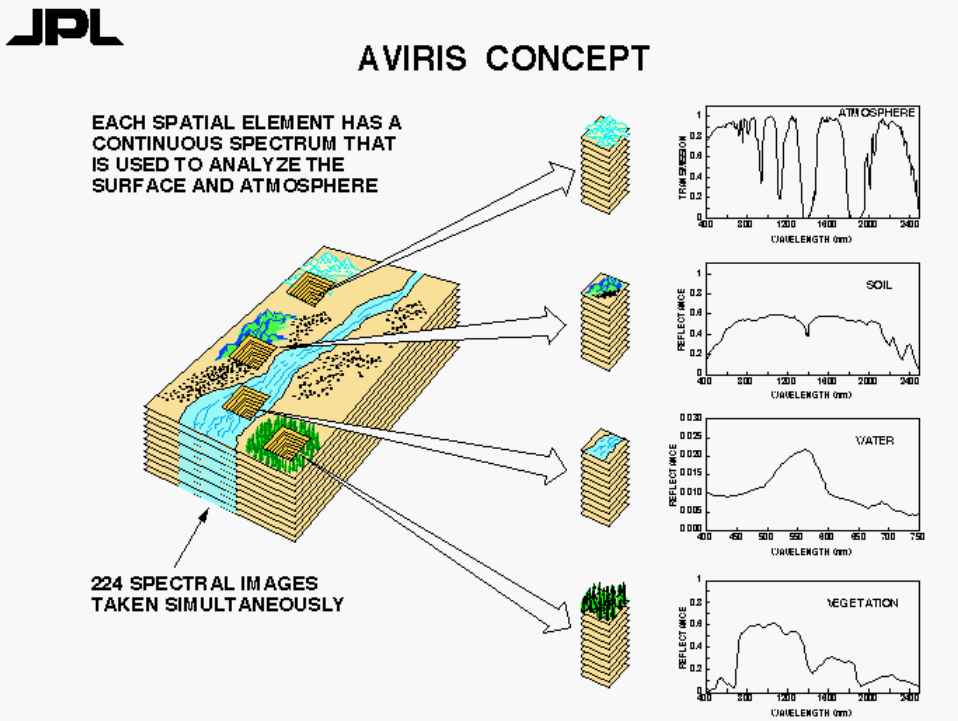

Figure 9: Imaging spectroscopy, or hyperspectral imaging, allows stand-off material analysis. This conceptual figure appears at [20].

Imaging spectroscopy has been used in both laboratory environments as well as remote sensing environments. Commercial applications for interior imaging or surveillance (similar to that of digital camera systems) have not been common due to the high computing and post-processing requirements of hyperspectral image cubes. Hyperspectral images contain not only two-dimensional spatial values, but also hundreds of spectral bands associated with each pixel location. As shown in Figure 9, a separate image can be shown in each spectral band, or spectral analysis can be performed at every pixel. Hyperspectral imaging provides powerful capabilities for analysis, and often is called remote chemistry.

A survey of current commercial spectral cameras returns several companies [21,22] that provide capabilities from the UV through LWIR. These companies list applications as material or chemical identification, color measurement, machine vision, gas and chemical detection, moisture profiling, and counterfeit detection. For safeguards, any of these applications could be considered. Another possibility would be the ability to purposefully embed “tags” into materials and use spectral cameras for tag verification [23], equipment authentication, or tamper-indication.

5. Polarization

Another optical field that is underutilized is polarization. Polarization can provide information about surface features, shape, shading, and roughness [24]. This information is largely uncorrelated with spectral and intensity images, and thus has the potential to enhance features.

One particular use for polarimeters has been for underwater imagery to mitigate the effect of scattering of light by the water. A polarization analyzer can be put in front of a digital camera and the polarization state can be adjusted to maximize the contrast between the object and background [24].

Figure 10: Reflection of cloud removed on water surface using polarizer filter.

In photography, polarizing filters are used to reduce reflections, as well as seeing through windows and water, as shown in Figure 10.

Polarimeters are also used in industry for sample characterization and evaluation. Specifically, polarimeters are used in the glass and sugar industry, the food industry, and in pharmaceuticals to look for chirality of molecules.

For safeguards applications, polarization might be used for tamper indication, equipment authentication, or for imaging through water, windows, high-reflection conditions, or where uncorrelated optical information is beneficial. There are many new research opportunities in polarization, and new applications and capabilities may emerge.

6. Conclusions

Imaging based on radiation from the visible band of the EM spectrum is one of many imaging technologies available. Furthermore, within bands of the EM spectrum, technologies are available to make use of other optical information, such as increasing the spectral resolution to enable imaging spectroscopy and utilizing polarimetric information. Even acoustic imaging, although not part of the EM spectrum, is an available technology for imaging applications.

These technologies are currently and commonly applied in fields other than safeguards. For instance, imaging spectroscopy is common in airborne geological surveys. Physical protection uses NIR illuminators for low-light applications. Thermal imaging is used in process control and now more recently by nuclear facilities for physical protection.

As resource management becomes more critical in safeguards, adaptation of technologies common to other industries could have the potential to provide improved technologies or even complimentary surveillance with less research costs funded directly by the safeguards community.

Not every technology in this survey paper will be applicable to safeguards. It is the hope, however, that this survey paper will provide enough background to spark interest and perhaps focused-research for safeguards applications in these technologies.

7. Acknowledgements

Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy’s National Nuclear Security Administration under contract DE-AC04-94AL85000.

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Exploration of Ion-Exchanged Glass for Seals Applications

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Abstract:

As the nuclear industry grows around the globe, it brings with it a need for more safeguards and proliferation resistant technologies. The International Atomic Energy Agency (IAEA) depends on effective containment and surveillance (C/S) technologies and methods for maintaining continuity of knowledge over nuclear assets. Tags and seals, a subset of C/S technologies, are an area where innovation has been relatively stagnant for the past fifteen years (pickett lecture). Seals are used to maintain the integrity of monitoring enclosures, containers, or perhaps a point of entry. Tags are used like barcodes, as unique identifiers to account for separate items. It is necessary to investigate technologies not previously used in this field in order to defend against emerging threats and methods of defeat.
Based on a gap analysis of tags and seals currently being used by the IAEA, completed with the input of several subject matter experts, the technology selected for investigation was ion-exchanged glass. Ion-exchanged glass is relatively inexpensive, has high strength, and can be used in a variety of applications. If identical pieces of glass are exchanged under the same conditions and subjected to the same point load, the fracture patterns produced can be compared and used as a verification measure. This technology has the potential to be used in passive seal applications.

Keywords: seals, verification, containment and surveillance

1. Introduction

As the nuclear industry grows around the globe, it brings with it a need for more safeguards and proliferation resistant technologies. The International Atomic Energy Agency (IAEA) depends on effective containment and surveillance (C/S) technologies and methods for maintaining continuity of knowledge over nuclear assets[1]. Tags and seals, a subset of C/S technologies, are an area where innovation has been relatively stagnant for the past fifteen years[2]. Seals are used to maintain the integrity of monitoring enclosures, containers, or perhaps a point of entry. Tags are used like barcodes, as unique identifiers to account for separate items. It is necessary to investigate technologies not previously used in this field in order to defend against emerging threats and methods of defeat.
Previously, a gap analysis evaluating the tags and seals the IAEA currently uses, along with technologies not developed for used in containment and surveillance applications was developed. This analysis utilized the input of several subject matter experts. The technology selected from this analysis for investigation was ion-exchanged glass. The research presented demonstrates the ability

to compare and match fracture patterns of identically ion-exchanged glass disks for verification purposes.

2. Background

The areas of containment and surveillance where the ion-exchanged glass can be applied are important to note and provide a framework for some of the design requirements a device incorporating the ion-exchanged glass may have to meet. Additionally, understanding the process of ion-exchange is necessary in order to take full advantage of all advantages associated with this process.

2.1. Tags and Seals

A seal is a tamper indicating device designed to leave non-erasable, unambiguous evidence of entry or tampering. The purpose of a seal is not to restrict or prevent access but just record that it took place.[1] Seals are mainly used for arms control and material containment, and therefore need field verification and authentication capabilities.
Tags are unique assigned identifiers or intrinsic features that are used for asset identification.[1] The purpose of a tag is to ensure that it is extremely difficult for an adversary to counterfeit an individual identification marker that is applied to, or inherent to an asset.[3] The IAEA uses tags to document individual assets and ensure that unauthorized replacements are not made.

2.2. Ion-Exchanged Glass

The process of chemically tempering glass, or ion-exchanging glass, is accomplished by immersing the glass in a molten solution of potassium nitrate where the Na^+ ions, close to the surface in the glass are replaced buy the K^+ ions from the solution. Figure 1 shows the process of ion-exchange. This process (ion-exchange) is thermally activated and results in the strengthening of the glass.[4,5,6] The increase in glass strength is dependent on the time and temperature at which the ion-exchange occurs.[7]

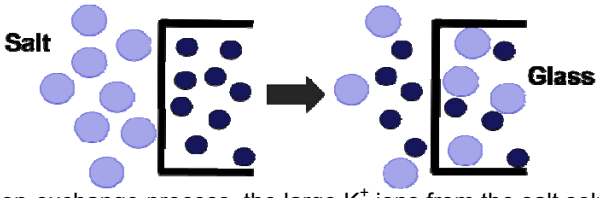

Figure 1. Schematic of the ion-exchange process, the large K^+ ions from the salt solution exchange with the Na^+ ions in the glass

Several papers and presentations concerning the fragmentation of ion-exchanged glass have been published and used as guidelines for the experimental procedures of this study. At present, two papers have been published discussing the fragmentation behavior and crack branching patterns found in ion-exchanged glass; however there has been no work done to match the fragmentation patterns of two identical pieces of glass that were ion-exchanged under the same parameters.[7,8]

2.2.1. Ion-Exchanged Glass Applications

There are several options for using ion-exchanged glass in tag or seal applications for C/S. The simplest application is to use an initially fractured piece of glass as a tag. This would require a piece of glass to be contained, its fragmentation pattern preserved, and the glass attached to an asset and photographed. At a later date the fragmentation pattern would be inspected and compared to the originally documented pattern.
Using ion-exchanged glass in a passive seal would be beneficial for securing the ends of a wire loop between two pieces of glass. The two ends could be sandwiched between two glass disks, of known ion-exchange parameters, and bonded by an adhesive. When the seal is removed from the asset each disk could be fractured and analyzed. This post-mortem inspection would rely on the ability to verify the fracture pattern produced by the disks and compare it to known standards for glass ion-exchanged

under the same conditions. Analyzing and verifying the fracture pattern will rely on image analysis which can easily be performed with basic equipment in a lab setting.

3. Fracture Procedure of Ion-Exchanged Glass

The glass used in this study was an alumino-silicate glass, Corning 2317 or Gorilla® Glass. The 2317 glass was ordered as a set of 20 disks, 5.08 cm in diameter and 2 mm in thickness, which underwent ion-exchange in a potassium nitrate bath for 48 hours at a temperature of 450°C (Marathon Glass, Stillwater, MN). The samples were ordered to these specifications in order to minimize possible discrepancies when following the experimental procedures outlined by Tandon and Kooi.

3.1. Experimental Setup and Procedure

The samples were cleaned with acetone and transparent tape was applied to one side of each sample. The samples were marked with a felt tipped pen in the center of the disk on the taped side. This mark was used as a reference for aligning the indenter tip with the center of the samples. The samples were fractured by loading the center of the un-taped surface with a Vickers macrohardness indenter (Buehler Macro Vickers 1900-2005) at 30 kg load, applied at a speed of 70 μm/s and held for 20 s. The tape held the sample fragments together after undergoing the indentation process.

4. Fracture Pattern Analysis

Upon preliminary visual inspection the fractured sample were divided into two groups, those with a ‘3 leaf’ fracture pattern and those with a ‘4 leaf’ fracture pattern. If the samples did not exhibit either of these patterns they were considered a failed sample. One sample was lost during the fracturing process, leaving 19 samples to be evaluated. Table 1 shows the fractured sample number and the respective number of leafs determined by visual inspection. Evaluating this data, based on a total of 19 samples 68.42% of samples fractured in a ‘4 leaf’ pattern, 26.32% fractured in a ‘3 leaf’ pattern, and 5.26% of the samples failed. Effectively, this grouping produced two samples groups, ‘3 leaf’ and ‘4 leaf’ fracture patterns.

Sample Number	Number of Leafs
1	4
2	Lost
3	4
4	4
5	4
6	3
7	4
8	Failed
9	4
10	4
11	4
12	3
13	3
14	3
15	4
16	4
17	4
18	3
19	4
20	4

Table 1. Fractured Sample Number and Corresponding Number of Leafs

4.1. Image Analysis

Image analysis of the fracture patterns utilized the Image Processing Toolbox in MATLAB. A code for image alignment and spatial transformation took advantage of the Control Point Selection Tool, allowing the user to interactively select points on a pair of images (MATLAB help). In each comparison one image is defined as a 'base' image while the other is the 'unregistered' image. The 'unregistered' image will undergo the spatial transformation for the control point pairs in each image to be aligned and is ultimately displayed with a semitransparent overlay of the 'base' image for comparison as seen in Figure 2. This image provides a visual reference but is not substantial for fracture pattern authentication or verification. It should be noted that fracture pattern comparisons were only performed with samples of the same group.

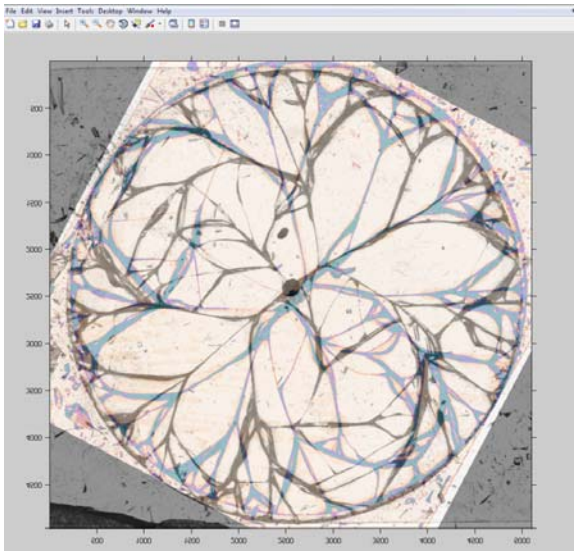

Figure 2. Semitransparent overlay of two images created using the MATLAB

The images are also analyzed in ImageJ, an image analysis software developed by the Research Services Branch of the National Institute of Health. The images can be imported from MATLAB, already overlaid, or can be manually rotated, translated, and overlaid. A desired pair of images can be combined using the XOR function, this function uses pixel addition displaying two overlaid black pixels as white. This means where two cracks are aligned white pixels will be seen. Thus, evaluating the number of black pixels compared to white, gives a percentage for determining how similar the fragmentation patterns are between two images.
This percentage of black to white pixels evaluation is achieved by defining a circular area of 0.200 inches (0.5 inch diameter) about the center of the resultant image. The analysis was performed on the central region of the images to avoid edge effects, where the sample underwent ion-exchange on three surfaces which produces a greater degree of fragmentation (Kooi paper p. 216).

5. Results

Basic statistical analysis, using the software Minitab, was done on the fractured samples. Probability plots, histograms, and other data characterizations were produced for both the '3 leaf' and '4 leaf' datasets. These characterizations provide a sound basis for deducing meaning from both datasets.
The '3 leaf' dataset had a total of 5 samples, providing 10 combinations of images; the data shown in Table 2 is a summary of specific characteristics. Among these specific characteristics are skewness which measures the lack of symmetry in data, and kurtosis which determines whether data is peaked or flat, relative to a normal distribution. These characteristics can be displayed using a histogram with a normal fit overlay, as seen in Figure 3. The same analysis was done on the '4 leaf' dataset of 13 samples, and 78 image combinations; the data is displayed in Table 3 and Figure 4.

Mean	16.181%
Standard Deviation	2.661%
Variance	0.00071
Skewness	-0.311
Kurtosis	-0.259

Table 2. Characteristics of '3 leaf' Composite Images

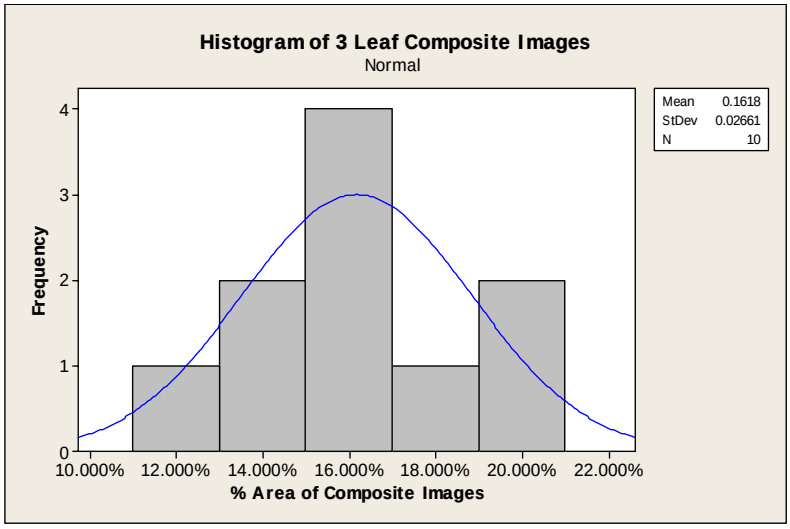

Figure 3. Histogram of '3 leaf' Composite Images

Mean	20.543%
Standard Deviation	3.448%
Variance	0.00119
Skewness	-0.251
Kurtosis	-0.695

Table 3. Characteristics of '4 leaf' Composite Images

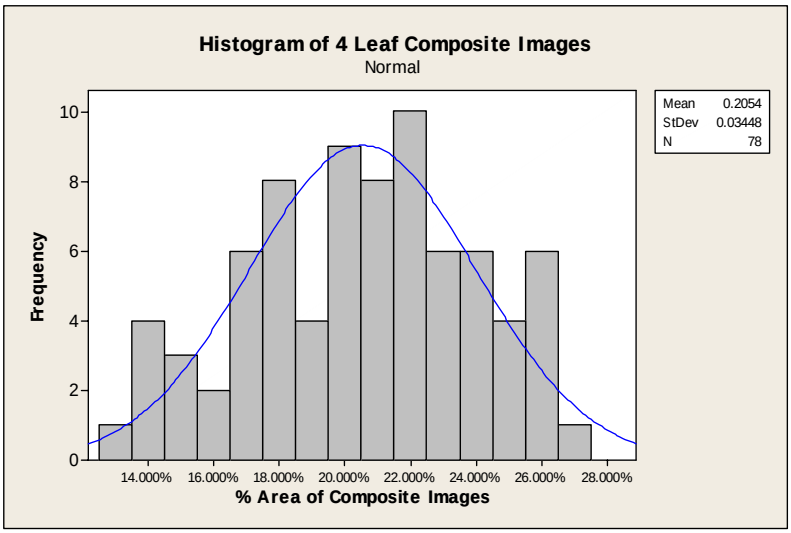

Figure 4. Histogram of '4 leaf' Composite Images

The histogram of the '3 leaf' composite images will not provide much useful data, given that the dataset is only 5 samples. The histogram of the '4 leaf' composite images displays a larger spread of data and it complements the overlaid normal fit. It is interesting to note that both data sets are skewed to the left and are considered flat relative to the standard normal distribution.

It is important to simulate the post-mortem verification technique for glass seals if they are deployed in the field. One method to simulate this analysis is to select a sample from the '3 leaf' or '4 leaf' populations; this will be called the 'field' sample. The samples not selected from the respective populations will be the 'control' samples.
The composite images created with the field sample and the control samples from the respective population were analyzed. The percentage of black to white pixels , or the% Area, for each composite image was computed and the average calculated. Composite images of the control samples from the populations were produced for all possible remaining combinations and the % Areas calculated and averaged. This process is repeated for each sample in the respective populations.
The difference between the averaged % Areas of the field and control samples was calculated and compared to the standard deviation (± σ) of the correlating population. If the difference is less than or equal to the standard deviation the field sample is considered a match to the controls, this is shown in Figures 5 and 6. The majority of the data points fall within one standard deviation, giving an 80% pass rate for the '3 leaf' fracture patterns and a 78% pass rate for the '4 leaf fracture patterns.

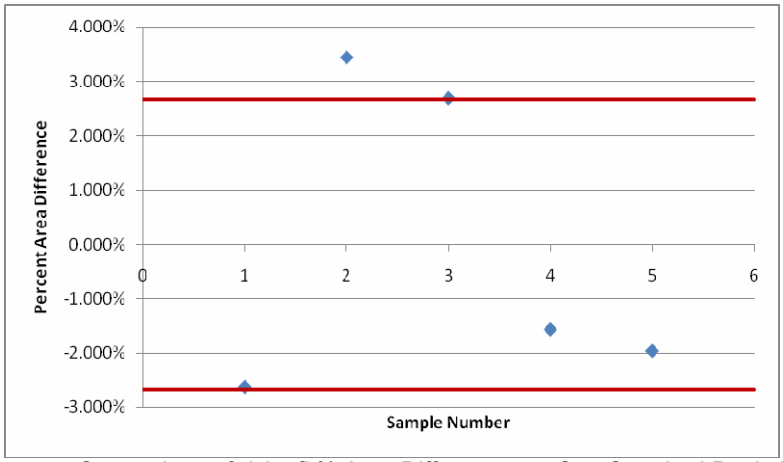

Figure 5. Comparison of '3 leaf' % Area Differences to One Standard Deviation

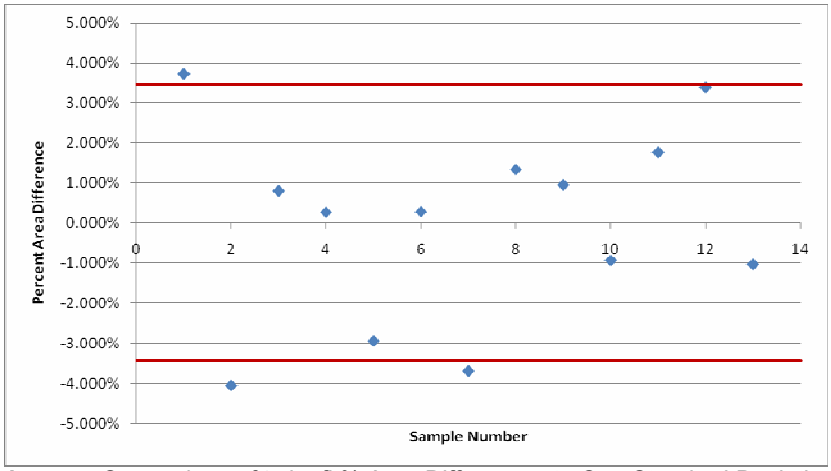

Figure 6. Comparison of '4 leaf' % Area Differences to One Standard Deviation

6. Conclusions

Based on the preliminary results it appears that ion-exchanged glass is a viable technology for a tag or seals application. Although more statistical tests need to be done pass rates of 80% and 77% for one standard deviation at a 95% confidence interval should not be discounted. A larger sample population needs to be assessed in order to validate these findings. Indeed further study necessitates another sample group that has undergone the ion-exchange process with different parameters to truly evaluate the possibility of verifying glass based on distinct fracture characteristics.

5. Acknowledgements

The authors would like to acknowledge Dr. Eric Taleff at The University of Texas at Austin for the use of his laboratory facilities, Dr. Stanislav Vitha at the Microscopy and Imaging Center at Texas A&M University for his time and expertise. In addition a special thanks and acknowledgment goes to Dr. Raj Tandon at Sandia National Laboratories for his expertise.

6 Legal matters

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Preliminary studies on remote inspection for JRC CANDUSealing Systems

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Abstract:

This paper presents the current implementation status of the JCSS (JRC Candu Sealing System): seals, reading system, seals database. The number of deployed seals and inspections is expected to grow in the next years, and there is a strong demand for a way to optimize the inspection procedure. One possible solution may be remote inspections, where an inspector is present in the initial sealing phase, but the following periodical verifications can be performed also by the site operator. The current system is designed to be used only by a nuclear inspector. Design modifications and improvements needed to achieve a safe and reliable remote inspection are analyzed. Conclusions on possible scenarios and future developments are provided.

Keywords: Spent fuel storage; ultrasonic sealing bolts; underwater seals; Remote inspection

1. Introduction

Nuclear spent fuel is kept in underwater storages under safeguards. The Joint Research Centre of the European Commission (JRC) developed an ultrasonic sealing system to seal stacks of spent fuel assemblies under water. The seals demonstrated a very good stability and reliability. The process to deploy the seal and to inspect them is a completely manual procedure and it is always done in the presence of an inspector. It is anticipated that the number of sites under control will increase. As a consequence there will be an increase in the number of seals deployed and the number of inspections. The required personnel resources for all the inspections can become very expensive.
The importance of remote safeguards inspection has been already highlighted by IAEA in [1]. Some parts of the industrial fuel cycle are fully automated, and studies to perform verifications with the help of remote monitoring techniques of such activities (L. Persson et al. [2]) have been published.
For spent fuel verifications, so far many verification activities are manual operations and need to be run by inspectors on site. It is therefore of great interest to have a system in which manual operations, such as in-field verification tasks, can be performed also by the personnel of the site-operator. Below we discuss a system which could allow significant inspection resource savings without affecting the authenticity and validity of the readings. The site operator could be allowed - under video surveillance - to collect data without the presence of an inspector. The collected data can be transmitted to and authenticated by the relevant authority and can be used for verification purposes

2. The JRC underwater ultrasonic seal verification system

The JRC ultrasonic sealing system is a perfect example in which a seal is placed on stacks of spent fuel assemblies and periodically verified to detect unauthorised activities. One application of the sealing system has been described recently (ref [3])

Figure 1 The JRC CANDU Sealing System and detail of the reading head

The verification procedure is currently carried out directly on site by an inspector.
The system consists of a PC for data processing, a box for data acquisition and a reading head with an ultrasonic transducer (Fig. 1) that is placed on the seal to acquire the seal ultrasonic fingerprint. All the instruments for the verification of the seal identity and integrity are always kept in a trusted environment and operated by inspectors. Since an adversary cannot have physical access to any element of the verification system, the only part that can be currently attacked is the seal. The seal design and manufacturing process ensure its strength against attacks as it was also confirmed in a vulnerability assessment performed by an independent laboratory. The seal is installed by an inspector according to a precise step by step procedure in order to avoid tampering. More details on the system can be found in [3].

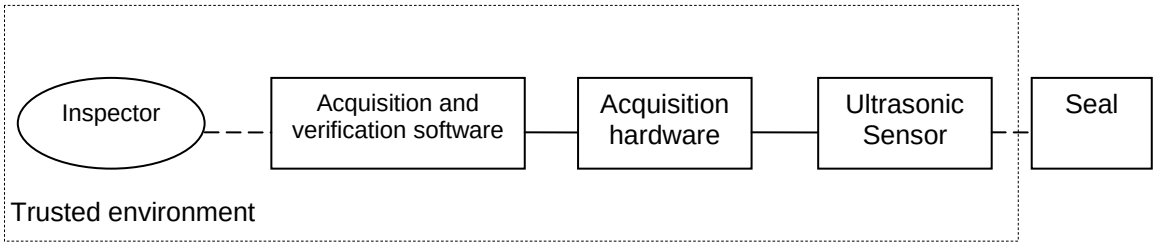

Figure 2. Current block scheme of the sealing system

In the current configuration, only a trusted and authorised person can do this operation, since he is the base of the physical security of the system.

3. Remote inspection scenario

In a future moving towards remote inspection, the current system must be adapted and enhanced to ensure the required security level. It is very important to highlight the fact that even if the seal can be remotely inspected, the installation procedure and the removal procedure will be always performed by an inspector. This is fundamental to ensure that the seal is placed on the item declared by the site-operator. Ultrasonic underwater seals are used to seal stacks with spent fuel that are stored for many

years, so normally the remote inspections are done in areas where surveillance camera are in operation. The possibility to perform remote inspections during the life time of the seal would bring benefits in term of time and cost to the overall sealing procedure.

3.1. New boundaries

In a remote inspection scenario the data acquisition is not performed by a nuclear inspector. Based on this assumption, the scheme proposed in Figure 2 is no more valid. In this new scenario, all the items are physically accessible by an adversary and all of them may expose vulnerabilities.

The first step that should be undertaken to convert the current system into a remotely controlled one is to split the "acquisition and verification software" into two separate blocks "Acquisition" and "Verification".
The "Verification" block contains all the sensitive information required for the verification, and will be still kept permanently in a trusted environment, such as the headquarters of the verification authority.
The "Acquisition" block contains all the technical parameters required for the readings of the seals, but does not contain any information on past readings, and does not provide any useful information for future readings or the verification algorithm. This separation is feasible, and has been tested in our laboratories.

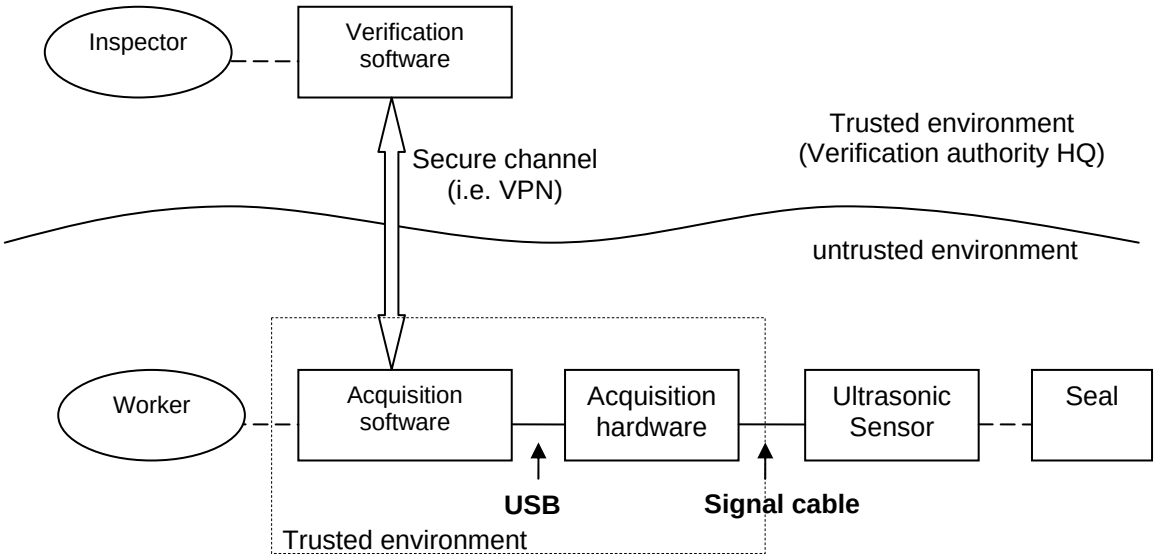

Figure 3 Online remote verification process

The verification process can be done in two ways:

- online, where the acquisition software is connected to the verification software and the data are sent in real time (streaming)
- offline, where the acquisition software is working stand-alone and the data are collected and stored in an encrypted file that is later sent to the verification software

3.2. The streaming approach

Assuming that the Acquisition software and the Verification software are connected by means of any kind of communication media, cabled or wireless, the process of verification of one seal can be done in two ways. One way is a pure "remote operation" mode where an inspector sends commands to the Acquisition software on the site and the on-field worker must do all the manual operations following the instructions from the Acquisition software.
A second way is to allow the on-field worker to interact with the Acquisition software, and do the data collection in a more autonomous way.
The benefit of the first way is that there is a strong control over the operation of the worker, because each step is followed by an inspector. The drawbacks are that the use of human resource is not optimal. Time differences between countries can also make this solution impractical.

The second way is much better from this point of view, giving the on-field worker more flexibility. Keeping a real-time connection between the Acquisition and Verification software ensures that no data is stored locally, making it more difficult to create false data. It also allows real-time controls, so the in-field worker cannot know if there is an inspector on the other side monitoring his activities. The actual presence of an inspector is not mandatory, since the Verification software may automatically collect the data sent by the in-field worker, and the inspector can perform the verification of all the collected data at a later time.

Even if it can be technically a good solution, a streaming approach can pose many problems in the implementation. In many sites data communication is missing or restricted for security reason. The system is strongly dependant on a data communication channel, so if it is not available or it is not reliable, the system will not work. For this reason off-line solutions only are evaluated in the following paragraphs.

3.3. The off-line approach

A different approach consists in having the in-field worker performing the data acquisition in a totally autonomous way. The system should be a very simple device that provides step by step instruction to a worker on the way to read a seal fingerprint and provides a clear visual signal when the seal is read correctly. The data are collected from the system and stored in an encrypted file that should be sent to the authority for verification. This very simple approach poses a number of problems concerning the security. In this scenario, only the file generated by the Acquisition software is available to the inspector in the verification phase. It means that the different parts of the system may be altered to provide false data.

The current system should be strengthened to reduce to the minimum the number of interfaces exposed to non-trusted personnel. The best way is to have one single box protected against tampering containing all the electronics and software needed for the acquisition, eliminating every interface that an adversary may attack, and leaving only the interface to the reading head. In this configuration (Figure 3) the security of the system can be split in three parts: security of the data sent to the verification authority, physical security of the acquisition box and security of the ultrasonic sensor.

The data security is a typical problem of data authentication and encryption. There is a lot of literature on the topic and it can be solved using well known IT practices. Most software authentication and encryption techniques rely on keys to be kept secret. In this case the problem is shifted to the physical protection of the secret keys. The box should have the necessary anti-tampering features that can ensure that the secret data is destroyed when tampering occurs. The same problem is being addressed by other security devices.

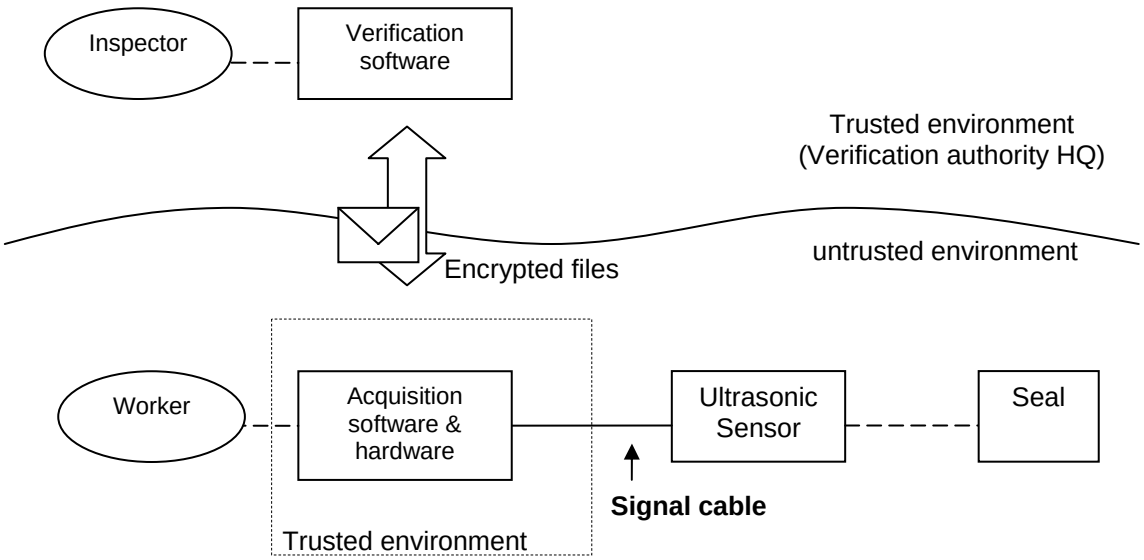

Figure 4 Off-line remote verification process

4. Vulnerabilities in an offline remote inspection scenario

As a foreword, it should be considered that in a seal verification scenario, missing data are treated like an indication of tampering. This means that in case an operator is requested to read a list of seals, an incomplete list of seal readings should initiate further investigation. From this assumption, attacks that will block in any way the data flow from the site to the verification authority are not considered.
Based on this assumption the proposed scenario is examined to highlight the main types of attacks that an adversary may try.

4.1. Replay attack

This attack is based on the capability from an adversary to get a reading from an intact seal, record it and send it to the verification software any time a verification is requested. The most known example is the video recorded from a security camera played back in a loop to hide the real images. In this scenario the acquisition software can use time-stamping and/or an authentication method with the verification software to avoid the replay attack on the encrypted data. In this way every reading of the seal will produce a different encrypted data file. A weak spot is left, and it is in the communication between the acquisition software and the ultrasonic sensor. It is not proved, but it can be imagined that a well motivated adversary may create an electronic device that will act as an ultrasonic sensor, with the ability to record a valid signal and replay it towards the acquisition hardware, even without knowing what it contains.

4.2. Man in the middle attack

In this attack the adversary places something between two points of communication to gain access to information and eventually modify it during the transit. In the remote inspection scenario we suppose a strong authentication system between acquisition and verification software. The connection between the Acquisition software and the ultrasonic sensor is again the weakest link, where an adversary would try to modify only the part of the signal that carries the proof of tampering.

4.3. Forgery (cloning)

This attack consists in creating a device that behaves like a reading head with an ultrasonic sensor but actually is a clone under control of the adversary.
Taking into consideration that the acquisition process is not a single reading but a set of different acquisitions with different parameters, these attacks seem very difficult to achieve. Anyway, they will be kept in consideration in future developments.

It should be taken into consideration also that there can still be some unannounced inspection, or the inspector may be on site for installation of new seal, and while using the equipment would detect the anomaly.

5. Results

From the analysis above it is clear that one of the main points is to have a reliable way to guarantee the authentication of the ultrasonic sensor, or, in other words, the reading head device as a whole.

The main problem is that the reading head carrying the sensor is working under water in a radioactive environment. The reading head cannot use complex electronics onboard, which may fail under radiation, so the instantaneous digitalization and encryption of the signal inside the reading head is not a feasible solution.

If we consider that the seals are kept under safeguards (with access control and monitored by cameras) the problem may be mitigated by a procedure that allows to authenticate the reading head in the area under an optical surveillance and then keep the acquisition system always under surveillance, to avoid swapping the reading head during operation.

All the attacks discussed are aimed at hiding an unauthorized break of seals. Assuming that the removal of the seal is always performed by an inspector, this action would be discovered when the seal is removed by an inspector, who will find a seal broken before removal. In case the adversary will tamper the equipment, the broken seal will read as not broken even after the inspector has removed it. This anomaly will be detected by the system.

6. Conclusions and future works

In this paper we presented the current sealing system developed by the JRC for underwater spent fuel sealing. The system is conceived to be operated by a nuclear inspector. Moving towards a new system that does not require the inspector to be present at the time of the verification of the seal is a big challenge. Some possible scenarios have been discussed and evaluated. Advantages such as time and cost efficiency and weaknesses due to an increased exposure to possible tampering have been highlighted. Some obstacles are still under study to be overcome.
The research on the physical security of the reading head will continue, studying new ways to authenticate the signal, new ways of tamper detection and new inspection procedure to obtain a remotely operated system that can ensure the same security level of the system used by inspectors.
Moving even further with the future possibilities of the system, once the security issues are solved, a field of research will be the full automation of the process using cameras, force sensors and a robotic manipulator.

7. Acknowledgements

The authors express their gratitude to all the colleagues who helped during talks, discussion, coffee breaks providing impressions and ideas. We would like to thank Joao Gonçalves for the useful information provided on surveillance. We would also like to thank colleagues from IAEA and DG ENER that provide us with valuable feedbacks and set always new challenges.

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"IAEA - EC joint remote data transmission"

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Abstract

The Nuclear Safeguards Directorate of the European Commission and the IAEA have jointly designed and successfully implemented a system for common remote transmission of safeguards information from the EU civil nuclear installations to their respective headquarters in Luxembourg and Vienna. This is a valid example on how the IAEA and the EC can make common use of technical equipment for implementation of modern safeguards in the EU.

Common IAEA-EC use of equipment for remote monitoring has faced serious challenges in finding appropriate technical solutions which should ensure at the same time:

- Fulfilment of the technical specifications for the requested performances (e.g. type and flow of data, transmission lines)
- Robustness against any potential attempt to tamper the transmitted data
- Secure handling of data complying also with operators' and National authorities' requirements
- Simultaneous access to the transmitted data for both organisations

Modern technology which has been recently made available as COTS, allowed the IAEA and the EC to find an elegant solution satisfying their common needs.

The system architecture, as well as the intrinsic security features, are presented and discussed.

Keywords: Data and information evaluation methodology, remote monitoring and secure data transmission

Reference Configuration for Reliable and Secured Data Acquisition and Remote Data Transfer

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Abstract:

The Directorate for Nuclear Safeguards of the European Commission has to upgrade most of its IT infrastructure in the large bulk handling facilities of the EU in the next years. With the Data Acquisition Infrastructure Monitoring and Management System (DAIMMS) project and a reference data acquisition infrastructure based on virtual PCs on redundant servers the Directorate intends to standardise the hard- and software as much as possible to simplify the maintenance, to improve the reliability and redundancy, and to increase the security of data transfer. The early and secured separation of state of health and safeguards data will allow a separate transfer of these data and also the use of external maintenance services. The concept in detail and first field test results will be discussed.

Keywords: Data and information evaluation methodology, remote monitoring and secure data transmission

1. Introduction

In the large facilities EURATOM Safeguards is using a lot of permanently installed safeguards equipment to follow up and verify the flow of material in processing areas and the inventories of stores. 'Large facilities' in this context mean facilities in which Pu (Plutonium) is handled in bulk form – for example the MOX (Mixed OXide) fuel fabrication facilities or the reprocessing plants in France and the United Kingdom. An overview of different systems, experiences and challenges is given in [1].

2. The Status of Safeguards Systems in Large Facilities Today

2.1. Used Hardware

Today's permanently installed systems consist in general of the following hardware:
- custom made or industrial sensors like NDA detectors together with related electronics and high voltage (HV) supplies (i.e. a shift registers (JSR) for neutron coincident counting, multi channel analysers (MCA) for gamma spectroscopy, serial multichannel counter (SMC) for a simple gamma, neutron or fork detector measurements), electronic seals, cameras, ID readers, pressure and level meters or outputs from other industrial devices (4…20 mA, voltages, switchers, etc),
- industrial PCs with interfaces like RS 232, 485 or CTC (Counter Timer Cards) and installed RADAR [2] software, which is recording the data and controlling the sensor concerning setup and check of operation status,
- network equipment like switches, routers, FO (Fibre Optic) converters to allow the authentic, authorised and accounted transfer of data and control of the sensors and PCs via VPN (Virtual Private Networks) from a central inspector office in the different installations and
- several PCs or servers for data storage, review and evaluation onsite in the central inspector office.

For plants sending data via RDT (Remote Data Transmission) systems to our HQ, as has been the case for the Sellafield site since 2007, a big part of the data evaluations can be done at HQ. Therefore

the efficiency of inspections will increase as the inspectors can concentrate on onsite verifications and the resolution of open questions.

2.2. Used Software

For data acquisition and control the RADAR/CRISP and OPC compatible software is used.

RADAR (Remote Acquisition of Data and Review) has been developed by EURATOM Safeguards since more than 13 years. It was designed for long term development and upgrade to have a standardised fully modular software platform for all safeguard related devices and RDT. With this software package we are able to acquire data and control the electronics of most of our safeguard devices like neutron and gamma detectors including their HV supplies, JSRs, multichannel analysers, etc including electronic seals and cameras. The DAMs (Data Acquisition Modules) are in charge of controlling the respective devices. If new devices like the JSR 15 should be controlled, only an appropriate DAM has to be developed. By using the module RadarLog the DAMs are able to create the output as daily files for folder based storages which we are using for small and stand alone applications like fork measurements as well as to fill an OPC compatible data historian database mainly used for large applications (see Fig. 1).

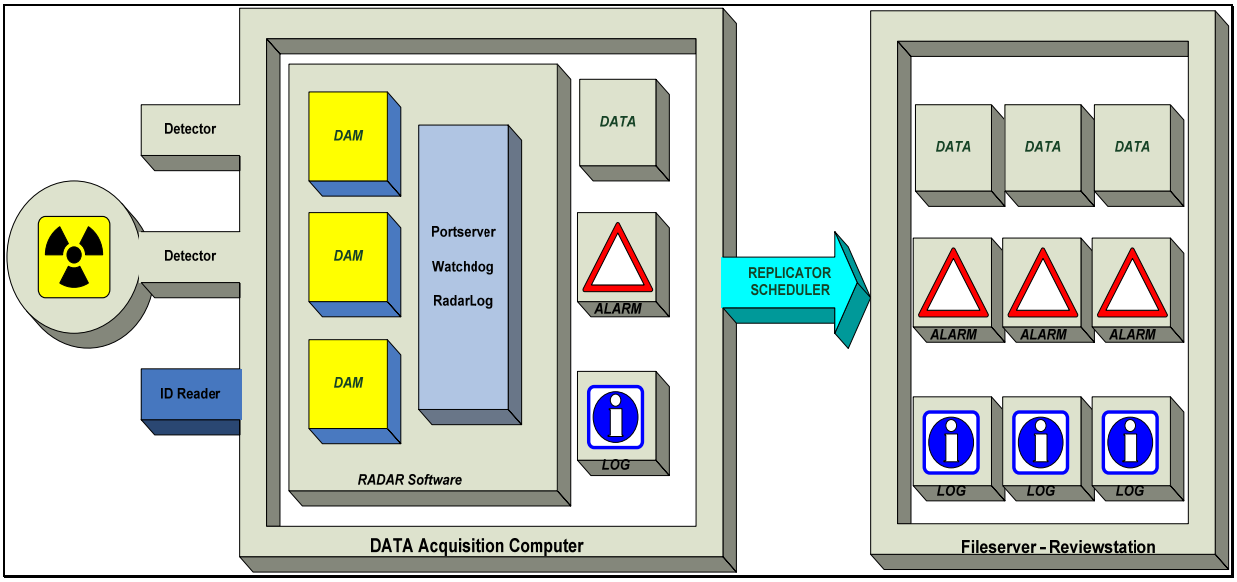

Fig 1: Unattended Data Acquisition by RADAR

CRISP (Central Radar Inspection Support Package) is a software platform to support inspectors during their review and evaluation of various sensors records. It can create events from continuously recorded RADAR files or data historians and allows a time based visualisation and correlation of different sensor events and records (see image 2). It is used mainly for event related measurements of itemised nuclear material i.e. the flow verification of Pu cans in reprocessing plants or pellet trays in MOX fuel fabrication plants. In cooperation with DOE laboratories, standard safeguards software has been modified and integrated into the CRISP package. Thanks to the available functionalities of MGA, ORELLA and INCC the operator declaration can be automatically compared with the value resulting from the unattended measurements. More details about RADAR and CRISP can be obtained from [2…4]

In the last 10 years OPC (Object linking and embedding for Process Control) has become a standard software for industry automation and is designed as a common interface for Windows based software applications and process control hardware.

The OPC compatible OSIsoft PI database with real-time data acquisition offers a central repository for data for a facility or across multiple locations. Information can be automatically collected from many different sources (Control systems, Lab equipment, Calculations, Manual Entry, and/or Custom software). Most information is gathered using one of the many OSIsoft and third party PI Interfaces. Inspectors can then access this information using a common set of tools (i.e. Excel, web browsers,

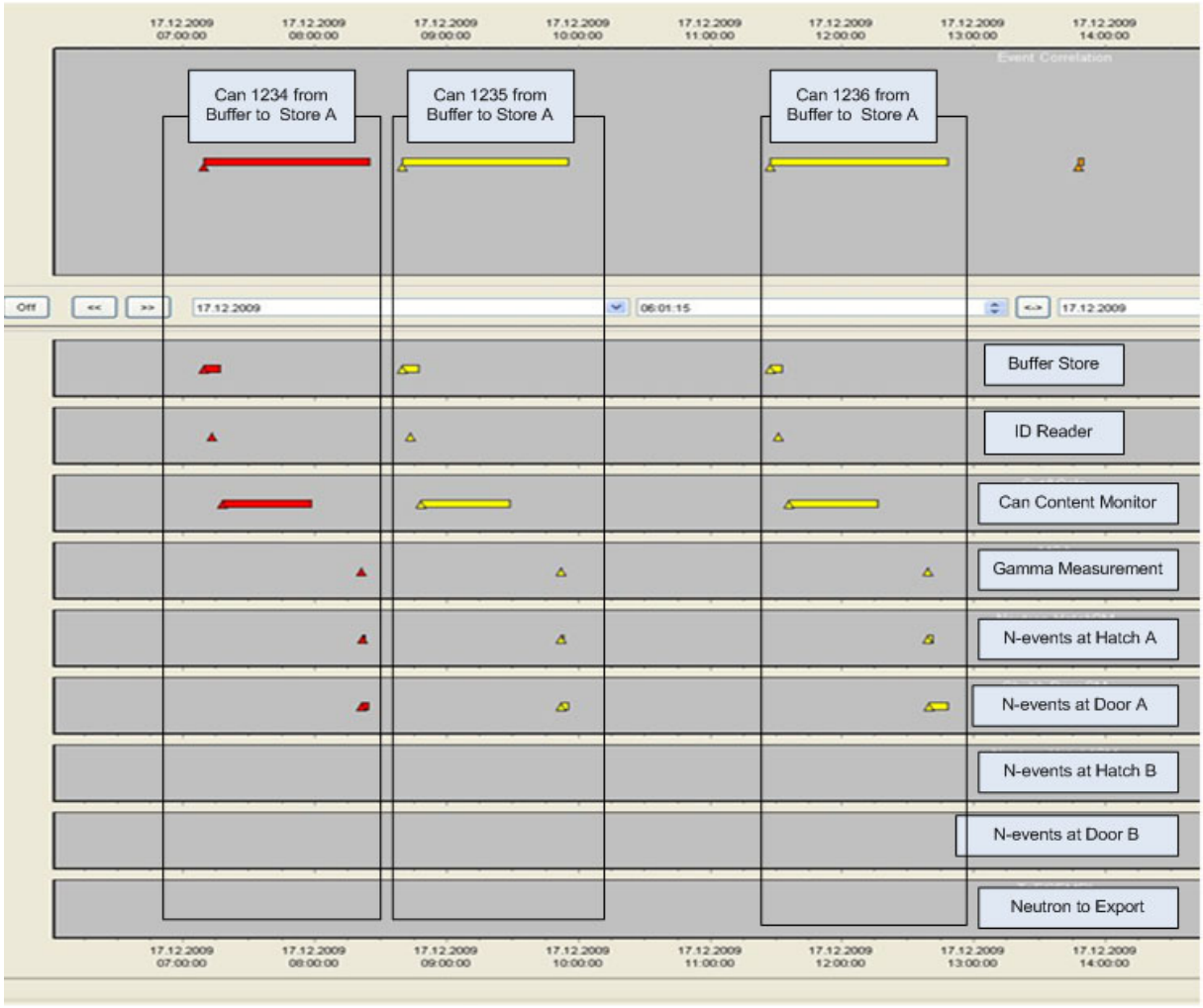

Fig. 2: Event visualisation of flow verification by CRISP – a click on the events makes the data visible

DAI (Data Acquisition and Interpretation) …) and look for correlations, analyze trends, determine if utilities are not meeting the demands and compare the performance of different lots of raw material. This software is mainly used for slowly changing values recorded by industrial sensors like pressure gauges, flow or tank level meters to allow flow verification of nuclear material in liquid forms in reprocessing plants.

2.3. Challenges for the upgrade

The number of different hard- and software systems used for safeguards purposes has grown over the years and continue to grow. Despite our effort to standardise the sensor electronics as much as possible there are now different generations of safeguards devices and electronics, PCs, data loggers or network equipment and different software versions in the field.

The IT hardware needs to be replaced periodically and a lot of equipment is between 5 to 10 years old which means that an upgrade is necessary soon.

The detector electronics, the PCs and part of the network equipment are often housed in 19" cabinets near to the detectors and therefore located in controlled areas, where access is limited and the work and maintenance on this equipment has to follow certain time consuming and expensive approaches.

The always increasing number of permanently installed systems needs more human and financial resources for installation, maintenance and upgrade, but these resources are limited.

Due to permanently increasing requirements of safety rules and fire protection obligations the status inside several cabinets are not state of the art anymore. An upgrade of these cabinets is necessary to respect the general rules and to make them easy to maintain.

Whereas the hardware currently is requiring more attention, the software will also need to be upgraded soon. RADAR and CRISP is based on Windows XP and a migration to Win 7 (or 8) has to be prepared and investigated. For more user friendliness some software parts have to be maintained and the automated evaluation has to be increased.
The raw data viewer ViewDAM was recently rewritten by state of the art software tools and is now rolled out under the name VisoR (Visualisation of RADAR data).
By incorporating the existing ORELLA burn up calculation software [5, 6] in CRISP we will soon be able to evaluate near real time fork detector measurements directly with declared operator values. Further tests are ongoing to extend the software capabilities for different spent fuel geometries and different data structures of operator declarations.

3. The Reference Configuration

3.1. The Goals

In order to address properly the hardware upgrading issues it was worthwhile to develop a concept for a Reference Configuration for Data Acquisition and Evaluation in large nuclear installations to resolve several challenges in one step. The goal of this concept was
- to increase the dependability of the systems by built-in reliability and redundancy or diversity,
- to reduce the maintenance effort and difficulties - minimise the use of equipment in the controlled area as much as possible and concentrate it in the onsite inspector offices,
- to standardise the hardware and the software as much as possible to increase the efficiency for maintenance and repair (especially storekeeping in HQ and onsite), to reduce the documentation effort and the training of inspectors to use this equipment and maybe help in replacements,
- to get near real time knowledge about malfunctioning of components by remote monitoring of the status of the systems and remote data transfer to HQ (including automated evaluation of alarm and log messages) to allow quick repairs or remote maintenance
- to enable (after detailed investigation) the replacement of faulty components with the help of inspectors or local contracted maintenance personnel
- to be prepared for a remote setup, reboot and configuration of all data acquisition and evaluation systems
- to have a maintenance and replacement plan for the remaining equipment
- to perform all upgrades with reasonable costs and to have reduced maintenance costs in the future.

3.2. The Concept

The concept was to install wherever possible 2 redundant and diversified lines of data acquisition in order to minimise the risk to lose data if one component in the chain fails. All parts located in controlled areas should be simplified and as reliable as possible. PCs should have a small size, be without a fan, have low power consumption, be rack mountable and the power supplies should be reliable and redundant. Complex software which will require frequent updates, fine tuning or comprehensive maintenance should be located on PCs in the inspector office.

Redundancy of sensors

For various reasons it is often not possible to double all NDA detectors completely (costs, space,…). But based on our early intention to increase the reliability a lot of installed detectors are already equipped with a redundant output (i.e. HPGe detector preamplifiers). Most of the neutron detectors are further redundant because in each of them the HV and power input and the preamplifiers are doubled and one half of the He tubes are connected to one group, the others to a second group. The pulses of each group are added and delivered to 2 outputs. If one of the detector components breaks the counting efficiency will be reduced, but in general the detector will remain operational.
In cases were other sensors are important for drawing safeguard conclusions (i.e. ID readers, seals, cameras or process sensors) it can be decided if a doubling is worthwhile or if a branch of operator signals is sufficient.

Redundancy and diversity of data acquisition and storage

The sensors are the starting point for the 2 redundant data acquisition chains (see Fig. 3). The primary chain or system will include the dedicated sensor electronics like several JSR, MMCA, SMC or a PC if pulses or operator signals have to be logged. To reduce the number of PCs for data recording and controlling of sensor electronics in the cupboard in the controlled area, it was decided to replace all

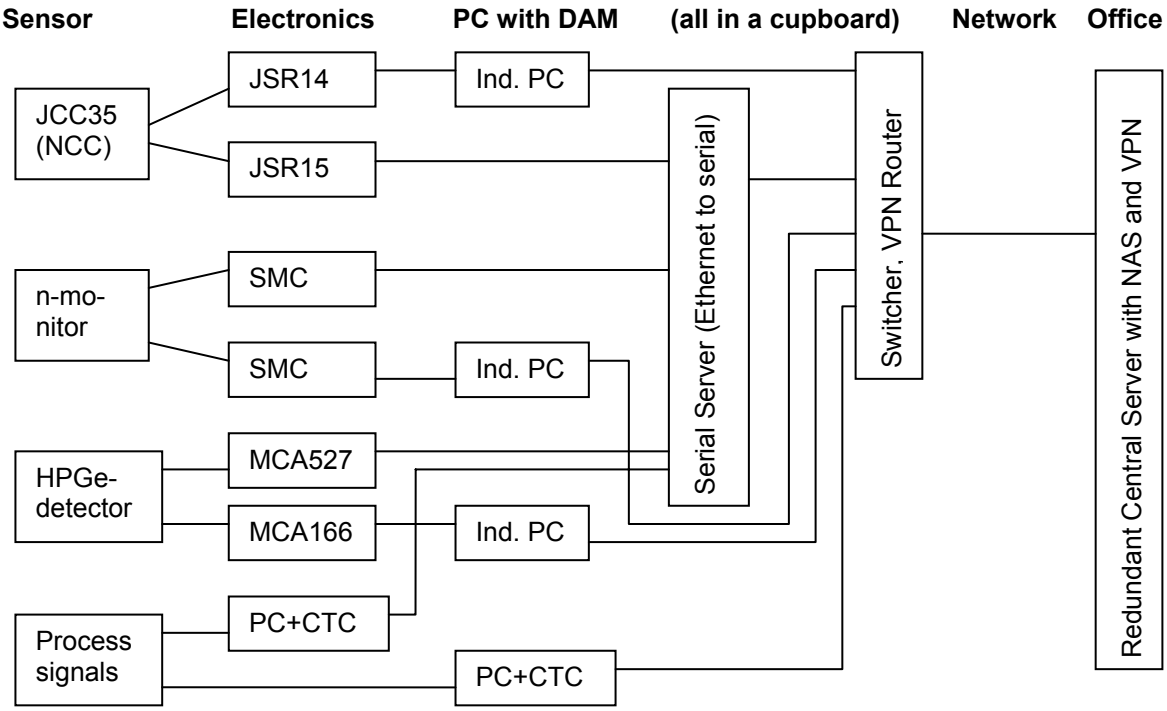

Fig 3: Example for primary (from electronics directly to serial server) and secondary data acquisition chain (via PC with DAM)

redundant PCs with RADAR-DAMs by a small and reliable serial server and control this serial server over a network from the inspector office, where a "hot" redundant Central Server with a NAS (Network Attached Storage) is located. On this Central Server several virtual PCs are running. Each of them control and record the data of one sensor electronics by using the appropriate RADAR-DAM. On the same server CRISP is also installed for onsite data review and evaluation. If one of the server breaks down the other will be automatically continue the job. The NAS system with built-in RAID system ensures that both servers use the same data set.
The critical path in this primary system is the network, because if the out of order time is longer than the storage capacity of the sensor electronic buffer it will result in a data gap. Therefore the network should be very reliable and also redundant. But the existing network cables cannot be doubled, because cable tracing in controlled areas will be too expensive. The redundancy can be achieved by intelligent switchers who are connected to each other by a ring of network cable. If a breakdown occurs on one side in the ring the switcher will automatically route the signals via the other side. This kind of network ring will be installed in Melox soon by adding cables between 3 rooms (see Fig. 4).

The secondary chain is a modernisation of the existing concept. All of the back-up sensor electronics is located in a cupboard and connected to a small, redundant powered and fan less industrial PC. RADAR with several appropriate DAMs is running on this PC to guarantee an autonomous data recording and independent storage on a SSD for several months in case of failures in the primary electronic or in the network and the redundant servers. If only the primary electronics or the serial server is out of order and the network and the Central Server continue to work the software could be remotely configured to use the stored data of the back-up system as an input for the CRISP algorithm. This means that a repair (or response) time of several days or weeks can be covered without a loss of data and/or performance.

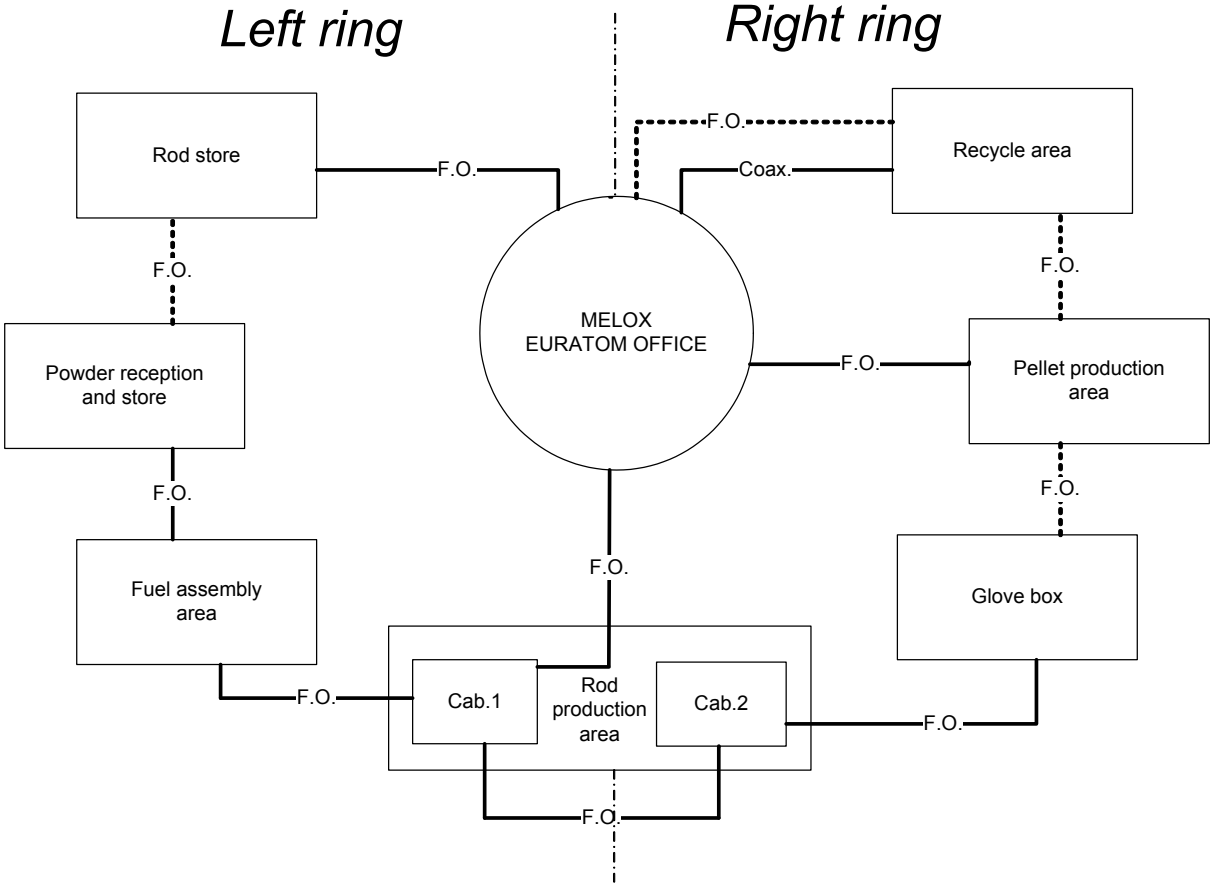

Fig 4: The future network in Melox (full lines: existing cables, dashed lines: to be installed)

Important for a quick response time is the near real time monitoring of the hardware in the system. The discovery time of an issue should be much shorter than the maximum response time before a loss of data will occur. This time depends on the buffer capacity of the different hardware. For a data recording the buffer time can be easily extended to several months by having sufficient buffer storage but for power failures the response time is often less than 1 week. Therefore the power supplies of the 2 measurement chains should be separated wherever possible, but never connected to the same UPS because power failures are the most likely failures.

The DAIMMS project

For the near real time discovery of failures the DAIMMS (Data Acquisition Infrastructure Monitoring and Management System) project was launched about 2 years ago. The aim was to develop a RDT monitoring system which is able to detect in near real time issues of data acquisition by very dependable permanent monitoring of the whole system. The system is built to grant high availability and with its embedded intelligent features it is capable to perform a continuous configuration control. This configuration control allows for easy configuration of components and provides an overview of complex system architectures, including the detection, localisation and documentation of the components of the whole data acquisition infrastructure. The visualization of the status of the different components and of their main configuration parameters completes the configuration control interface. The repeated checks of configuration and an unattended evaluation of the SNMP traps, generated by each device, are admitting automated documentation of changes and the remote localisation of problems related both to the status of devices and to the access of safeguard data. For the visualisation of the results "Castelrock" or NAGIOS can be used, depending on how flexible the system needs to be in the future. It will be possible to see from HQ how the DAMs and the network in Sellafield are configured, if the UPS (Uninterruptable Power Supply) batteries are in good conditions and if the systems are running stable.

Advanced implementation of RDT will not only allow for more efficient inspections; it will also give the possibility of remote maintenance including diagnostics, preventive maintenance and bug fixing [5]. The DAIMMS also supports an early and safe separation of monitoring and safeguards data. Groups of users which have only access to the monitoring data cannot access the safeguards data and vice versa. That will enable us to route the monitoring data also to a local maintainer in the large facilities, so that they might be able to fix certain issues without our presence. This idea has to be investigated further before it will be implemented.

3.3. The Implementation

The implementation depends on the urgency of replacement needs, the progress of the different tests and the activities in the installations concerned. The implementation can be separated in 4 steps:

- the tests of the virtualisation of the DAMs on Central Server
- the test of the DAIMMS project including visualisation of monitoring data
- the selection and purchase of hardware (serial server, DC power supplies, UPS and PCs) and
- the detailed plan for each project including network infrastructure

The test of the Central Server is ongoing on a mock-up in HQ. Two redundant servers and a NAS were installed, virtual PCs have been created and different safeguard sensors and their electronics have been connected via a serial server. The electronics is controlled by the appropriate DAMs and the whole system is in test currently. The preliminary in-house test results of the primary chain are very promising. By centralising the computer power on 2 servers in the inspector offices we think that not only the access for maintenance and upgrade will be easier and less time consuming, also the hardware costs will be lower as in cases where all PCs are doubled in the controlled area.

For the implementation of DAIMMS a generic prototype was developed and a Proof of Concept of a very sophisticated architecture was performed last year. The next steps are start of operation of the Network Operating Control centre for the main components installed in the remote Sellafield site as well as in the HQ machine and review rooms and to get practical experiences in:

- configuration of the SNMP suites for all sensors and electronics already offering these features,
- collecting SNMP information from RADAR functions and from the SoH (State of Health) logging files generated by other systems, such as SDIS, FAST, UPS and others to complete the coverage of monitoring by controlling all devices and functions running at the data acquisition chain,
- visualisation and presentation of the SoH monitoring results,
- configuration of network systems to grant high availability, confidentiality, integrity and security of data,
- ensuring strict and "impenetrable" separation between the flow of safeguards data and the flow of SoH monitoring information from remote devices and
- collecting exception reports and consequent accounting of information for further analysis about the access to devices and safeguards data as consequence of:
    - physical modifications of the infrastructure or
    - the attempt to violate secure transmission channels or
    - attacks against the authentication – authorisation mechanisms

In parallel to the remote 'SELLAFIELD' tests, we want to implement the above mentioned features also in MELOX as a "local" configuration where the system will support the local activities endorsed by Euratom Inspectors and will locally register events in a continuous, but (as long as RDT is not in place) not in real time, manner.

The selection of the important hardware components is almost finished. As an industrial PC a small fan less low power version with external 12V power supply with sufficient interfaces and plug-in extensions was selected. The missing fan is reducing the contamination risk extremely and the external power supply allows us to implement a redundant 12 VDC power supply per cupboard with one input from operator mains and one from internal UPS. To avoid common failure modes the primary system must be powered from a different UPS compared to the secondary system. Concerning the redundant 12 VDC and UPS module EURATOM Safeguards has a lot of experiences from former video systems. By consequent avoiding of common failure modes a very high MTBF (Mean Time Between Failure) was achieved.

The first detailed project plan was established for Melox. The network will be extended and upgraded as described in Fig. 4. The work might be finished by the end of the year. In the next weeks and months step by step all the cupboards in the different rooms will be upgraded following the above described concept. The secondary chain will be installed as described above. As long as the network is not completed and the Central Server is not installed the primary system will be a mirror of the secondary system and use the existing industrial PCs younger than 5 years. A challenge will be that during the upgrade activities the full functionality of the recording systems must be guaranteed because the upgrade will be made during continuous production. Once the structural basis for implementation of the above described primary system will be implemented (maybe by the end of this year) then the old PCs will be replaced by a serial server per cupboard and the full system will be ready for use.

The detailed project plan for a second place will be established soon. Based on the experiences from Melox we want to upgrade SMP in a one shot approach,

4. Summary

The large number of different data acquisition systems used in the field, together with the necessary periodical upgrade of a lot of these systems made it worthwhile to develop a concept for a reference configuration, which is state of the art from both the hardware and software point of view. This will allow for a better standardisation, earlier failure localisation and easier maintenance, less training for use and trouble shooting and results in a cost reduction in the future due to higher availability of data so that an increasing number of systems can be handled by the same human and financial resources.

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13 Export Control

Impact of the entry into force of the Lisbon Treaty on EU Export Control Regime

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Abstract:
The entry into force of the Lisbon Treaty in December 2009 have had and will have a substantial impact on the EU export control regime.

The first major impact concerns the new role granted to the European Parliament in the decision making process. Acting as a co-legislator with the Council, the European Parliament clearly intends to be proactive in the field.

The second major impact is related to the representation and the participation of the EU and its Member States to the different international export control instruments and conferences.

The proposed contribution will analyse and present the two groups of potential consequences and how it has been implement by the EU and its Member States since the entry of the Treaty.

Keywords: export control; Lisbon Treaty; co-decision

Export control on dual use goods in the European Union

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Abstract:

As recalled by the EU WMD Strategy and the New Lines of Actions for combating proliferation, export control on dual use goods is an important barrier against proliferation, together with nuclear security and nuclear safeguards, to which it is linked through the Additional Protocol (INFCIRC 540). This requires export declarations for fifteen nuclear activities listed in Annex I, according to the list of controlled items foreseen in Annex II, derived from the NSG Trigger list.

The export control legal framework in the European Union is set by Council Regulation 428/2009, which sets out the general framework for EU-wide controls on exports, brokering and transit, and introduces controls on intangible technology transfers as foreseen by UN Security Council Resolution 1540.

Enforcement by customs is ruled by the Community Customs Code and its recent security amendments, founded on the Commission Decision on establishment of common risk criteria and standards for security and safety risk analysis. This is applied since 1 January 2011, and introduces a strong legal mechanism for the equivalent application of customs controls and treatment of the legitimate trade (AEO) at the EU external borders. It is aimed towards identifying high-risk consignments / goods that could have serious implications on the security and safety of the EU and its citizens, and contains a set of common risk criteria to be applied in the Member States' automated risk analysis systems in order to continuously screen advance electronic cargo information for security and safety purposes.

The present paper will provide an overview of the export control process in the EU, underlining the main issues and challenges.

Keywords: Export control, authorisation, European Union, international regimes, customs, enforcement, Green paper, Pool of Experts, Additional Protocol

1. Introduction

The history of proliferation in the 80's-90's has shown how the illicit transfer of dual use technology has been the key element allowing the development of competences and capabilities in the WMD area.

Export control started during the Cold War in the 50's with COCOM (*Coordinating Committee for Multilateral Export Controls*) [1]. It later developed in the early 70's following the entry into force of the NPT with the Zangger Committee (1971) [2] and the Nuclear Suppliers Group (NSG, 1975) [3], later followed by the other export control regimes: Australia Group (AG, 1985) [4]; Missile Technology Control Regime (MTCR, 1987) [5] and Wassenaar Arrangement (WA) [6] in 1996.

The control lists produced by all these regimes include dual use items, whose importance together with

“personal” illicit procurement networks was brought to evidence by A. Q. Khan’s and subsequent cases [7].

The lists, complemented by the chemical precursors included in the Chemical Weapons Convention, have been integrated into the European export control regulation since the 90’s and inspired also the US Commerce Control List (CCL), which has the same structure and coding.

Export control and nuclear safeguards developed in parallel, as two intimately linked barriers to proliferation. After the discovery of undeclared proliferation activities in Iraq, the introduction in 1997 of the Model Additional Protocol [9] to the Comprehensive Safeguards Arrangements [8] enabled the IAEA to gain access to a much wider range of information and locations.

The Model Additional Protocol also requires export declarations of “Trigger list” items [10] listed in its Annex II, related to nuclear activities listed in Annex I. A paper on the AP experience of some ESARDA members is part of the export control session of the symposium [11].

2. The EU export control legal framework

The European Union WMD Strategy of 2003 [12] and the subsequent New Lines of Actions for combating proliferation [13] clearly stress that export control on dual use goods is an important barrier against proliferation. At the same time, dual-use export controls have a very prominent economic dimension as export control decisions have a direct impact on trade flows and competitiveness.

Dual-use export controls, as part of the EU's Common Commercial Policy, are an exclusive competence of the European Union. The entry into force of the Treaty of Lisbon on Dec. 1, 2009 has strengthened the role of the European Parliament in the export control field by making it a key player in the adoption of EU legislation in the area.

The common framework on export control in the European Union is formed by:

- Council Regulation (EC) No 428/2009 of 5 May 2009 setting up a Community regime for the control of exports, transfer, brokering and transit of dual-use items [14]
- Community Customs Code – Council Regulation (EEC) 2913/92 [15] and its 2005 Security amendments [16]

EU Regulations are directly applicable in the European Union. Member States are responsible for their practical implementation. National security exceptions are allowed on a case-by-case basis.

3. Export control on Dual Use goods: main points of Regulation 428/2009

The framework for export controls in the EU is set out by Regulation 428/2009, which includes the following main elements:

- it foresees 4 types of export authorisations (EU General Export Authorisations, National General Export Authorisations, global export authorisations, individual export authorisations);
- it foresees controls on export transactions as well as on brokering and transit, as required by UNSCR 1540 [17];
- it covers intangible transfers of technology (ITT) including via internal IT systems;
- it sets out the general criteria that need to be taken into account when deciding whether or not to authorise an export;
- it foresees various consultation and information exchange procedures, including in particular concerning denials;
- Annex I contains a list of all items subject to export controls. The Regulation also foresees the cases where Member States may impose controls on non-listed items through so-called 'catch-all' controls ('end-use' controls);
- Annex II sets out the EU General Export Authorisations currently in force;
- Annex IV lists the items that are subject to intra-EU transfer controls.
- The Regulation allows Member States to extend controls in certain specific cases.

Whereas export control legislation is developed at EU level, it is the Member States that have the primary responsibility for the practical implementation of the controls. It is hence the Member States who issue export authorisations and denials, and who enforce the controls.

3.1 Annex I

The basis for the identification of items subject to export controls is the EU list of dual-use goods set out in Annex I of the Dual-use Regulation. As said in the introduction, it constitutes a consolidated version of the control lists agreed by the international export control regimes, plus the chemical precursors derived from the Chemical Weapons Convention.

The structure of Annex I is derived from WA's one, divided into categories numbered from 1 to 9, with the subsequent structured integration of the other regimes' control lists. Category 0, containing all the NSG Trigger List items, completes the organization of the annex, while NSG Dual Use items [18] are contained in the other categories.

Each entry contains a set of criteria and parameters which decide whether a particular item is subject to export controls or not. Only Category 0 items are mostly illustrative in nature, including the defined term "especially designed of prepared for nuclear use" derived from NSG terminology, which brings about that items should possess specific design features and labelling in order to be controlled.

Annex I contains about 1500 items and is regularly updated following the evolution of regimes' controls. As said, the Regulation also foresees the cases where Member States may impose controls on items not listed in Annex I through so-called 'catch-all' controls ('end-use' controls). This is the case of items that are judged to be also potentially useful to a proliferation programme, haven't been included in any regimes' control list yet, but may appear in so-called "watch-lists".

Annex I is adopted in many countries outside the EU, and was also the basis for the US Commodity Control List of EAR, which has exactly the same structure and items coding, but contains about 25% more items.

3.2 Annex II

Annex II is reserved to *EU General Export Authorisations* (EU GEAs): at the moment only EU001 is included, authorising the export of most dual-use items to 7 countries (Australia, Canada, Japan, New Zealand, Norway, Switzerland and the USA). EU GEAs facilitate export transactions as they eliminate the need to apply for individual or global licenses, which often take considerable effort. Each country's administration sets up instruments for verification of the correct use of these licenses.

In 2008 the European Commission put forward a proposal to introduce six new EU General Export Authorisations. These new authorisations should be put in place in 2011.
Some countries have National General Authorisations (NGA) in place which are similar in nature to EU General Export Authorisations, but cover different items and destinations, and are only available to exporters in the issuing country. A broader number of EU GEAs would enable exporters in all EU-27 to benefit of the same facilitated export conditions.

3.3 Annex IV: Intra-EU transfers

While Annex III contains export authorisation templates, Annex IV is a subset of Annex I listing those items for which an export authorization is requested also for intra-EU transfers.
It is divided into Part 1, including items for which National General Authorisations (NGA) are allowed, and Part 2, mainly encompassing Category 0 Trigger List items and other most sensitive ones, for which NGAs are excluded.

Intra-EU transfers are a paradox of the European Single Market. Whereas the free circulation of most dual-use items is ensured within the EU, items listed in Annex IV are still subject to an authorisation requirement when being transferred from one EU Member State to another. This requirement is the consequence of the historical national security origin of controls, which are now covered by the Common Commercial Policy.

Various reasons continue to be cited for why intra-EU transfer controls should remain in place including the need to fulfil international obligations with regard to reporting of transfers. Intra-EU transfer controls however continue to be a burden for EU multi-national suppliers which lament encountering more difficulties than competitors from other countries. There may hence be scope for certain reforms in this area.

3.4 Sanctions

Some Council Regulations adopting restrictive measures (embargos) against countries or non-states actors contain also lists of dual use items, additional to Regulation 428/2009 Annex I. This is e.g. the case of the current Regulation 961/2010 against Iran [19] and Regulation 1283/2009 against DPRK [20]. These regulations also include lists of entities to which exports cannot be authorised.

Sanctions are dealt by the Foreign Policy Instruments (FPI) service of the European Commission.

4. EU Export control enforcement: Customs Code and amendments

As anticipated in par. 2, the legal basis for enforcement is the Community Custom Code Reg. 2913/92 with its 2005 Security Amendments introducing key elements like Common Risk criteria for targeting shipments and requirements for information sharing.

The Customs Code and implementing provisions require establishment of an equivalent level of customs control in the Community to ensure a harmonised application of controls based on commonly defined risk criteria and standards for entry (including transhipment) and export/export operations.

the so called 'security' amendments to the Code introduced in April 2005 made a further step in strengthening the role and responsibilities of the customs authorities in providing security and safety of the EU by introducing three main elements.

1. The status of Authorized Economic Operator (AEO), which is made available to those economic operators who fulfil strict compliance requirements in securing their supply chains. In return these operators should benefit from the EU-wide recognition in terms of fewer customs checks and faster (border) clearance.

2. A Common Risk Management Framework (CMRF) ensuring equivalent identification of the illegal trade EU-wide and harmonised treatment of the AEO with specific focus on the security risks.

3. The requirement for advance electronic notification of the arrival and departure of goods at the border of the EU customs area was introduced to support automated security and safety risk analysis prior to arrival and departure of goods and implementation of the EU risk management framework.

Following these amendments, the Commission (Directorate for Taxation and Customs Union – DG TAXUD) and the Member States launched various legal measures and IT projects to implement these new requirements.

The effectiveness of the common risk criteria is constantly monitored and evaluated, to support a common approach and to ensure that priorities are set effectively and resources are allocated efficiently, with the aim of maintaining a proper balance between customs controls and the facilitation of legitimate trade.
Common risk criteria are defined to assess non-proliferation risks and to support the enforcement on dual use export controls by Customs.

The electronic EU *Customs Risk Management System* (CRMS) was set up to ensure an effective exchange of information among Member States Customs Administrations on identified risks.

Furthermore IT systems were established at the EU and national level to support the communication of the advanced cargo data from trade to customs, and the AEO programme implemented.

Along with information exchange, common and high standards are in place to support the efforts to detect nuclear, explosive, biological, chemical and radiological cargo especially through appropriate deployment and use of technical equipment like scanners and other Non intrusive inspection techniques (e;g installation of detection portals).

The full implementation of the security amendments in all EU-27 started on 1. January 2011.

5. Relevant EU committees

Export control implementation issues are discussed in “Article 23 Coordination Group” meetings, chaired by the European Commission Directorate General TRADE.
In addition to the *Article 23* meetings held in Brussels, a series of peer-visits are organised each year and are hosted in various EU Member States. Each Peer Visit meeting targets one specific subject of implementation. The host administrations set the background in agreement with the Commission, and chair the discussion. Proposed amendments to the legal framework on export controls are discussed at the Council’s “Working Party on Dual Use Goods (WPDU)”, chaired by the Rotating Presidency of the EU.

Since the entry into force of the Lisbon Treaty, the European Parliament plays a key role in the adoption of export control legislation by co-decision. The European Parliament's International Trade Committee (INTA) prepares the detailed position of the EP on export control issues.

“Customs 2013” is a co-operation programme and a forum for discussion among EU-27 customs authorities and Commission’s DG TAXUD. The programme runs for six years (Jan. 1st 2008 – Dec. 31st 2013) and provides the legal and financial base for:

- Reinforcing security and safety within the Community and at the external border;
- Strengthening the fight against fraud and protecting the financial and economic interests of the Community and Member States;
- Increasing the competitiveness of European business by speeding up customs procedures partially through the creation of a European paperless electronic customs environment.

Customs 2013 does not explicitly include dual use goods export controls, however joint workshops with Art.23 CG are regularly organised.

6. EU export authorisation implementation issues

Due to varying reasons, EU Member States have different resources allocated to export controls.
Due to the free circulation of goods within the EU, any of the 27 EU Member States can potentially be an exporter of any dual-use item. Consequently, it is of common EU interest that the whole system functions properly.
An analysis of measures taken to implement export controls in the EU Member States suggests that there are differences in the approaches taken by particular Member States.

6.1 Administrative procedures

There are different administrative procedures in place in the various EU Member States (e.g. registration requirements for exporters and reporting). Some Member States seem to require their exporters to have internal compliance programmes before being eligible to export dual-use items, while others do not.

6.2 Authorisations

Member States make different use of the various authorizations available under the Dual-use Regulation. For example, a few Member States have implemented broad National General Export Authorisations thereby facilitating exports for their exporters, while exporters from other Member States do not have access to such facilitation measures.

6.3 Operational differences

There are operational differences also due to resources and capabilities. Member State have different interpretations of control list entries and make different use of the 'catch-all' provisions of the Dual-use Regulation, allowing for the imposition of authorisation requirements on items not listed on the EU control list.

6.4 International regimes and access to information

Not all EU-27 have access to information shared within the international export control Regimes.
8 MS are not members of the Missile Technology Control Regime (Estonia, Cyprus, Latvia, Lithuania, Malta, Romania, Slovenia, Slovakia) while Cyprus is not member of the Wassenaar Arrangement.

This fact does not allow them to access information and authorisation denials issued by non-EU members. The denials issued by EU countries are shared within the DUES system of DG TRADE (see par. 7.3).

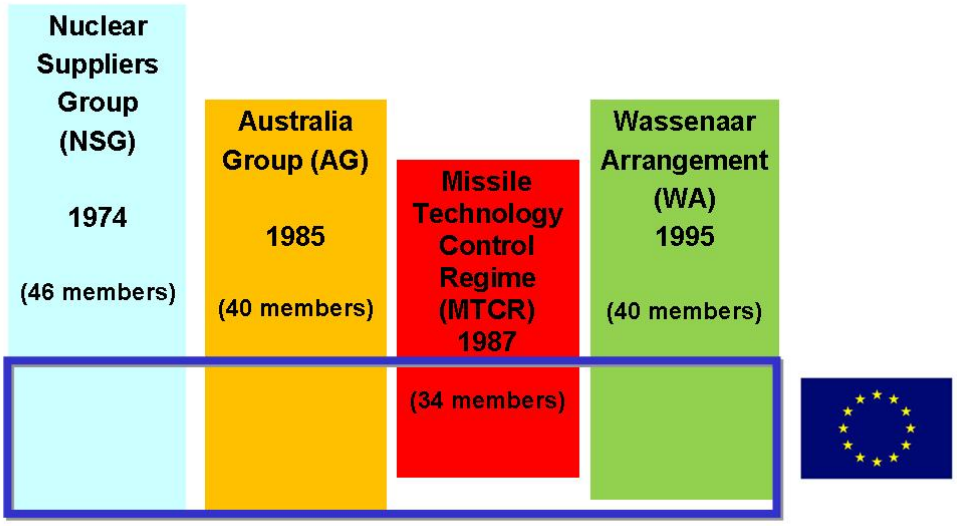

Figure 1 – EU-27 membership in international export control regimes

7. Improvement activities

7.1 Guidelines for implementation

The “Article 23 Coordination Group”, chaired by DG TRADE, is about to define Guidelines for harmonised interpretation and implementation of DU goods export control Regulation 428/2009, which should help to clarify part of the presented issues.

7.2 Green paper

To collect comments on the EU dual use export control system and views for the future, a *Green Paper* has been prepared by DG TRADE and opened for comments from all EU Stake-holders in the period July-October 2011 [21].

Article 25 of the Dual-use Regulation requires the Commission to prepare a report on the implementation of the EU export control system and possible areas of reform. Consequently, the objective of this Green Paper is to launch a broad public debate concerning the functioning of the current EU dual-use export control system.

The results of the consultation will therefore help identify the strengths and weaknesses of the current system and map out a longer-term vision of the EU export control framework. These results will be translated into concrete amendments to the current system and the preparation of a long term strategy on the development of export controls in the EU.

7.3 The Dual-use e-System (DUES)

Electronic systems provide a great help to exporters and authorizing officers.
Some EU member states have electronic licensing systems while others are planning to develop them in the near future. At a later stage, all of them could be sharing a defined amount of information.

The Commission launched on 26 January the Dual Use e-System (DUES), accessible to Member State

export control authorities. The system allows Member States to exchange export control information including in particular denials.

7.4 Technical expertise - Pool of Experts

The export control of dual use goods needs technical expertise to decide on the applicability of Annex I of Regulation 428/2009 and amendments. This interests licensing and enforcement, and possibly other phases of the process like prosecution.
However, technical expertise is not always available in all EU MS export control administrations, for lack of resources and specific knowledge.

To provide a broader access to expertise, an EU "Pool of Experts" on Dual Use goods was informally set-up in 2005, with experts made available by some EU member states and the Commission. In order to reinforce it, the Pool is now being reshaped to provide a more structured but light approach.
A proposal by the Commission JRC is being finalised in the relevant committees and foresees:

- A central point of contact (POC) for requests of expertise established by COM JRC (*pool-of-experts@jrc.ec.europa.eu*), which will allow a neutral follow up and monitoring of needs and resources, while maintaining the possibility of direct bi-lateral exchanges.
- A confirmed Pool of Experts: Commission experts will play a neutral central role to process the requests internally before forwarding part of them to national experts. The present "Pool of Experts" list will be refreshed.
- Allowed users: Requests of expertise will be accepted exclusively if formulated by officials of EU-27 export or customs administrations or related public offices. Advice will not be provided to private exporters.

The requests can relate to the control of dual use goods falling under EC Regulation 428/2009 and annexes. The advice provided will be non-binding, leaving the exclusive final decision to the national public administrations.

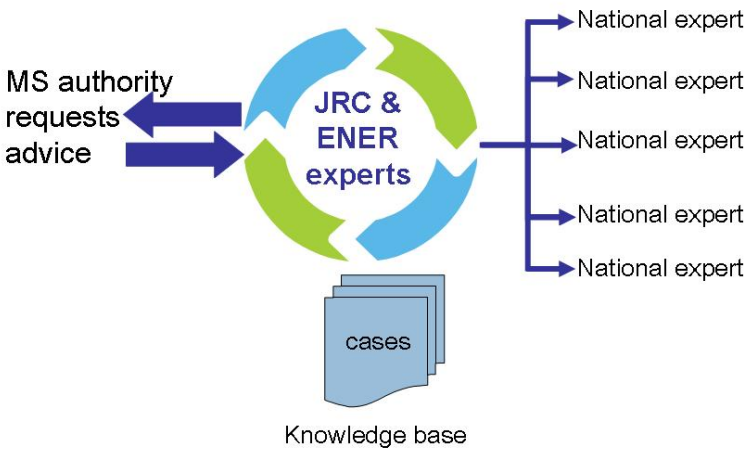

Figure 2 – Pool of experts on Dual Use goods

7.5 EU Inreach Training on Dual use export control

Linked to availability of expertise, the Commission is about to launch a programme on EU inreach training on dual use export control. The programme will be based on the results of an external study, which has outlined the needs and possible modules.

8. Conclusions

The European Union export control system is a key barrier that is set and developed to counter proliferation, as part of the European WMD strategy. Its implementation is a quite complex exercise, which must take into account the EU Common Commercial Policy and Customs Union, the national prerogatives of EU-27 and at the same time keep abreast of continuously evolving proliferation scenarios and threats.

As summarised in the paper, various initiatives are on-going to strive for a harmonised and effective implementation and enforcement of controls.

The feed-back to the Green Paper will define how the whole system should evolve and the new steps to be taken.

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The National Implementation of Nuclear Export Controls: Developing a Best Practices Model

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Abstract:

The nuclear renaissance promises significant benefits to the international community, but also raises security challenges, particularly relating to the trade of nuclear materials and equipment. The objective of this paper is to examine how supply-side non-proliferation efforts can be strengthened by developing a best practices model for national nuclear export control implementation. In order to achieve this goal, nuclear export control measures identified by the 1540 Committee will be used as a framework from which a best practices model can be formed. Such a model concentrates specifically on national legislation and enforcement measures delineated by the Committee in order to bring countries in accordance with international law. Developing a best practices model seeks to deliver an ideal process for national export control law actualization in order to encourage the peaceful development of nuclear energy and develop the infrastructure and framework for precluding nuclear proliferation.

Keywords: nuclear export controls; nuclear law; national implementation; nuclear non-proliferation, UNSC Resolution 1540

1. Introduction

The nuclear renaissance promises significant benefits to the international community, but also raises security challenges, particularly relating to the trade of nuclear materials, equipment, and technology. The objective of this paper is to examine how supply-side non-proliferation efforts can be strengthened by developing a best practices model for national nuclear export control implementation. In order to achieve this goal, it is necessary to examine first the elements international nuclear law, particularly focusing on UNSC Resolution 1540, that inform in detail specific national export control measures, and the national implementation of export controls, concentrating specifically on national legislation that has been brought about in order to bring countries in accordance with international law. Developing a best practices model delivers an ideal framework for national national export control implementation in order to strengthen international nuclear non-proliferation efforts.

The recurring issue emerging throughout discussions related to nuclear export controls constantly links shortcomings in the national implementation of international nuclear export control

standards to weaknesses in the international community's efforts to effectively use such controls as a non-proliferation tool. This paper's objective further derives from the desire to fill an evident literature gap, especially in academia, on the subject of national implementation of nuclear export controls. This gap is not present due to a lack of national jurisprudence but from a dearth of academic analyses of these laws, especially on a comparative level. That said, more or less descriptive research has been conducted on the export controls of specific countries, such as Pakistan and India, to name just two of the many.[1] Further, many think tanks and other institutions provide information regarding national export control laws. Most notable are the University of Georgia's Center for International Trade and Security and the Stockholm International Peace Research Institute (SIPRI). Additionally, several government and non-profit agencies have published so called " model" export control laws. Finally, it is important to mention that though few in number, the subject of nuclear export controls is gaining the attention of an increasing number of academics. What is currently lacking, notwithstanding the fine quality of these mentioned sources, is a cohesive study of a) an overarching best practices model related to national export control implementation b) a comparative study of export control laws in not just export control regime member states but new nuclear states c) an examination of not just export control laws but enforcement and compliance of violations, especially in new nuclear countries. This paper is an attempt at covering the first of these questions.

The methodology used for this study is as follows. First, many countries have public documents related to their export control laws. These can sometimes be found on ministry websites and legal databases, and in the instances where they are published, provide an excellent primary source. It is not surprising, however, that the majority of states do not have their specific laws published on ministry websites, at least as of 2011. Therefore, one of the most valuable sources of information regarding specific national laws are the UN Resolution 1540 Committee matrices and national reports. UN Resolution 1540 commands an international legally binding commitment from all states to implement measures in order to prevent non-state actors from acquiring weapons of mass destruction. While the Resolution deals with terrorism and relates broadly to chemical, biological, and radiological weapons, as well as their means of delivery, for the purposes of this study the principle point is that states must implement national legislation providing for nuclear export controls in order to preclude terrorist WMD attacks. States, according to paragraph 4 of the Resolution, must " present a first report no later than six months from the adoption of this resolution to the Committee on steps they have taken or intend to take to implement this resolution."

While not all states met the required deadline, by 2011 almost all have submitted a report. Expectedly, these reports vary widely in length, specificity, and quality. They consequently provide a rather good lens into the status of national implementation of export controls. Working from the reports, laws are examined, patterns discerned, gaps revealed, and even the shortest and most poorly written reports indicate a level of compliance that can be judged. It is also obvious that the reports

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[1] See Galhaut, Seema. "*Indian export control policy: Political commitment, institutional capacity, and non-proliferation record*." Center for International Trade and Security Issue Brief, May 2006.; or Pracha, Sobia Saaid, "*Strategic export controls: Case study of Pakistan.*" South Asian Strategic Stability Institute Brief, October 2009.

cannot be relied upon as a sole primary source for this study, but rather as a tool. The reports construct a path from which laws can be studied, grouped, and evaluated. In addition, the matrices evaluating national nuclear export control measures for each country provide a useful tool into examining exactly what specific steps are necessary in order to comply with the Resolution. Of particular interest are the last two sections of the matrix: measures related to OP 3 (c) and (d) and related matters from OP 6, 7, 8 and 10.

The paper will be presented in the following way. The first part presents the logic behind using UNSC Resolution 1540 as a guide for developing a national nuclear export control implementation model. The second part will address the measures necessary in order for a country to comply " appropriately and effectively" with the Resolution in terms of nuclear export control implementation. The conclusion will discuss problems and weaknesses related to the implementation of such measures, and offer conclusions offering insight into how the best practices model delineated in Part II can be better implemented in order to strengthen nuclear non-proliferation efforts.

2: UN Resolution 1540 as a Guide for National Implementation

Imagine a fictitious developing country in 2004, one that has immediate and grave problems, and few resources with which to solve them. It could be large or small, with ports or without, but for the sake of the scenario, let's say it has no civil nuclear energy program or domestic resources that could be traded for use in a nuclear program. This country, in 2004, is given the obligation by the United Nations Security Council to draft export control legislation for nuclear materials and equipment (as well as other WMDs), and in six months provide a progress report.[2] Even to a country that trades nuclear materials and equipment and has some kind of civil or military nuclear structure, the task of complying with Resolution 1540 is daunting enough. According to a study conducted by Princeton's Woodrow Wilson School, in 2007 no country was fully compliant, and " even the countries considered leaders in nonproliferation efforts would be no better than 50 percent compliant." [3] It is no small task, therefore, for our fictitious country to write its compliance report in the sixth month time frame given to it by the Resolution, in addition to actually creating, updating, or reforming its domestic nuclear export control laws.

Resolution 1540 is the strongest, and one of the only, pieces of international law mandating countries to implement national nuclear export controls. The Security Council had good timing, though, as it brought attention and urgency to the issue of export controls right at the beginning of the nuclear renaissance. The nuclear renaissance increases the trade of nuclear materials and equipment due to the increasing number of countries seeking civil nuclear energy programs, and this increased activity

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2 Some ambiguity exists regarding the state requirement to report, based on the wording of Operative Paragraph 4 which calls on states to report. See Craft, Cassidy. "*Brief challenges of UNSCR 1540: Questions about international export controls*." Center for International Trade and Security Briefs, March 2009.

3 "*A new urgency for non-proliferation: Implementing United National Security Council Resolution 1540.*" Princeton University Woodrow Wilson School of Public and International Affairs Graduate Policy Workshop Final Report, January 2006. 2007.

demands greater legal control.[4] The Resolution remains the single and most important international law obliging states to enact nuclear export control laws. Whether the Resolution realizes its objective, however, depends on how thoroughly states effectively implement it.[5]

The Resolution itself is extremely broad. First, regarding the context of the present paper the Resolution presses outside the scope of simply nuclear export controls by referring to chemical and biological, and not just nuclear, weapons. The usage of the umbrella term WMD achieves the goal of bringing greater attention to the terrorist threat of using such weapons to the international community, yet the practical consequence of using the term likewise multiplies the difficulty in implementing national laws that protect against all three types of threats. The risk presented by the divergence of nuclear materials and equipment is different from that of chemical or biological substances, and logically different laws should apply to each type of threat. This differentiation, however, is never enunciated in the Resolution, and therefore leaves the question of how to deal with the three- separately or together- up to states.

The Resolution is broad also in terms of what it expects from states. The broader the command, the more difficult it is to implement in practical terms, and the more room there is for a kind of fudging on the part of states if they wish to do so. Further, the Resolution does not address the consequences of non-compliance, such as possible enforcement actions or sanctions, giving states leeway in terms of the extent to which they implement the resolution. In fact, after the passage of the Resolution, one concern was whether states would actually submit their reports, as the strongest language in OP 4 is " calls upon."[6] In cases of non-compliance, the Security Council would be hard-pressed to impose sanctions or other punishment due not only to the lack of authorizing language in the Resolution, but by the inevitably staunch opposition such actions would face.

In terms of what the Resolution calls upon states to do, the text calls for the adoption of what in practical terms is quite a large body of law. Paragraph 2 calls on states to " adopt and enforce appropriate effective laws which prohibit any non-State actor to manufacture, acquire, possess, develop, transport, transfer or use nuclear, chemical or biological weapons and their means of delivery, in particular for 3S/RES/1540 (2004) terrorist purposes, as well as attempts to engage in any of the foregoing activities, participate in them as an accomplice, assist or finance them." The paragraph is relevant to nuclear export controls especially in the " acquire", " transport" and " transfer" mandates. However, many states find the wording of the text, especially of the use " appropriate and effective," too vague.[7] Surely " appropriate and effective" in this case means

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4 As of March 2011, the future of the nuclear renaissance has been questioned due to the events surrounding the Fukushima nuclear plant in Japan. However, notwithstanding several short-term political measures implemented in several countries, most plants in construction, or in planning, as well as civil nuclear cooperation deals, are continuing as envisioned pre-Fukushima.

5 Heupel, Monika. "Implementing UN Security Council Resolution 1540: A division of labor strategy." *Carnegie Papers*, No. 87, June 2007.

6 Craft, Cassidy. "*Brief challenges of UNSCR 1540: Questions about international export controls*." Center for International Trade and Security Briefs, March 2009.

7 Bergenas, Johan. "*A piece of the global puzzle*." Stimson Center Report, December 9, 2010.

drafting laws with the objective of precluding WMD proliferation, yet due to the complexity of such a large task, many states have expressed confusion as to what is concretely expected of them.

OP 2 by itself would only require countries to implement export controls in the context of terrorist end-uses. OP 3 makes no mention of terrorism and instead refers to proliferation in general, calling upon states to " take and enforce effective measures to establish domestic controls to prevent the proliferation of nuclear, chemical, or biological weapons and their means of delivery, including by establishing appropriate controls over related materials and to this end." Further, the Resolution, in OP 6, rather vaguely calls upon Member States, " when necessary", to implement control lists, because of their " utility." This is not therefore mandatory, but rather up to the interpretation of states. OP 8 then declares another rather strong obligation for states to fulfill. The paragraph directs states to do whatever they have to do to comply with international law in the context of non-proliferation, and additionally, in subparagraph (d), to " develop appropriate ways to work with and inform industry and the public regarding their obligations under such laws." This last part of the OP 8 is one of the most important parts of the Resolution for export controls. The simple existence of domestic laws, and even mechanisms for their enforcement, make little sense if they exist in a vacuum. Yet returning to the practical task of implementing the Resolution, it is clear that it is no small feat.

3: Using the 1540 Committee Matrices to Develop a Best Practices Model

3.1: The 1540 Committee Matrices- Preliminary Measures

The 1540 Committee has developed its own implementation matrices as of 2005 which organize information based on submitted 1540 reports. These matrices display how utterly complex it is to implement the Resolution. Taking the nuclear non-proliferation part of the Resolution by itself, there are dozens of measures to be taken by states in order to comply, especially in the area of nuclear export controls. The Committee states that the matrices are not a tool for implementation or measuring compliance with the Resolution, but rather " a reference tool for facilitating technical assistance" and " [enhancing] dialogue with States." Nevertheless, they serve as a rather useful instruments for gathering a list of what an effective national export control system looks like. Developing a best practices model for specific nuclear export controls stems from delineating what measures do in fact form an effective system. This section will therefore discuss the various measures listed in the matrices from a best practices point of view, informed by national law, national 1540 Committee reports, and other sources.

Before discussing the measures, it is worth remarking that the 1540 Committee matrices shed light on the vague wording of the Resolution, especially in areas where words such as " appropriate" and " effective" are used. In terms of the implementation of the Resolution, one difficulty countries have faced is simply understanding what exactly is meant by such language. As Peter Crail notes, " what is considered to be an appropriate and effective legal mechanism varies between states,

thereby complicating any such assessment and leaving room for political considerations to come into play as states assert that they are in compliance." [8]The matrices offer insight into what the Committee means by its language because they list the components necessary to comply with the separate parts of the Resolution. They are therefore a useful tool for understanding and organizing the varying measures necessary for the implementation of the Resolution, and for establishing a basic rubric for the domestic implementation of nuclear export controls in particular. While the matrices list measures rather straightforwardly, they do not explain how such measures are implemented in reality. Therefore, here the elements are discussed in detail, providing a guide from which states' actual implementation of the elements can be assessed.

Seven worksheets of the matrices correlate with different operative paragraphs of Resolution 1540 and for the purposes of this study, only the parts relating to nuclear export controls will be examined. The first worksheet discerns international treaties that reporting states have signed as well as stated commitments to non-proliferation, as related to OP 1 and related matters from OP 5, OP 6, OP 8 (a), (b), (c) and OP 10. From this worksheet it is therefore possible to identify the international commitments to nuclear export controls states have made, in particular by their adherence to the Non-proliferation Treaty (NPT), Convention of the Physical Protection of Nuclear Materials, participation in IAEA activities, and adherence to other treaties. It is a clear first step of any national nuclear export control system to be a party to principle international legal instruments, not only as a signal to the international community of a national commitment to nuclear non-proliferation but also because international law must then be implemented, and therefore acts as a model for domestic law.

Then next worksheet related to nuclear export controls identifies measures related to OP 2 of Resolution 1540. The paragraph is dedicated to non-proliferation in general, calling on states to " prohibit any non-State actor to manufacture, acquire, possess, develop, transport, transfer or use nuclear, chemical or biological weapons and their means of delivery, in particular for terrorist purposes, as well as attempts to engage in any of the foregoing activities, participate in them as an accomplice, assist or finance them." Especially germane to nuclear export controls are the " acquire," " transport" and " transfer" verbs in the paragraph. The prohibition of financing of proliferation activities is likewise key in this paragraph, as it has been noted that illegal nuclear transfers often take place with third party financial help. Identifying steps countries can take to comply with OP 2, the 1540 Committee matrix lists whether national legislation and enforcement is in place for the various parts of the paragraph, asking specifically for the source law involved in implementation and enforcement.

Needless to say, implementing the prohibition of activities listed in OP 2 in domestic law is not a simple task. The matrix does not offer insight into what such laws should look like, apart from the expectation that they cover the activities noted. It is an additional challenge for states to determine whether to group WMD activities together in domestic law or to treat chemical, biological, and nuclear activities separately. The 1540 Committee matrix separates the three activities into separate

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8 Crail, Peter. "Implementing UN Security Council Resolution 1540: A risk-based approach." *Non-proliferation Review, Vol. 13, No. 2, July 2006.*

checklists, but uses the same list of measures for each, thereby suggesting that domestic law should not group the activities together even if the laws are written in similar ways.

The worksheet related to OP 3 (a) and (b), which calls upon states to " develop and maintain appropriate effective measures to account for and secure [WMD and their means of delivery] in production, use, storage or transport" and " develop and maintain appropriate effective physical protection measures," lists criteria states must take to be in compliance with this part of the Resolution. In terms of relevance to nuclear export controls, this part identifies whether states have set up a national regulatory body, for example, as well as whether states have taken measures to account for the transport, storage, protection, accounting, use, production, and other activities that manage the movement and use of nuclear materials. Nuclear export controls, after all, require proper safeguarding measures to account for nuclear materials, as well as a system regulating safety while in facilities and during transport.[9] The IAEA uses the term " cross-cutting relationships" to demonstrate that other areas of domestic law, such as law regarding safeguards and physical protection, affect export controls. In this respect, it is important for states to implement domestic law stemming from international obligations under IAEA agreements and international nuclear law germane to OP (a) and (b).

The matrix also provides a worksheet related to OP 3 (c) and (d) and related matters from OP 6, and OP 10, and the activities listed here address directly export control activities and will be analyzed here in great detail. OP 3 (c) and (d) list " effective border controls and law enforcement" as well as " national export and trans-shipment controls" while OP 6 and 10 call upon states to act to prevent illicit trafficking and create national control lists. There are 26 distinct components noted in the matrix; these can be broken down into border control activities, licensing activities, legislation, enforcement activities, control lists, and funding/infrastructure measures.[10] These components of nuclear export controls illustrate the complexity of successful implementation.

3.2: Border Controls

To begin with the first group of measures listed, border control activities, it is necessary to understand how such activities relate to nuclear export controls. Some literature on Resolution 1540 refers to " border and export controls" as if border controls are not a part of export controls, but rather simply related.[11] The term border controls as used here, however, is used to signify an export control activity. After all, any cross border movement involves an import or an export of goods, services, or individuals, and monitoring such movements at inter-state borders forms just one facet of controlling such trade. This discussion opens up a further definitional dilemma: if illicit nuclear trafficking is defined by the IAEA as " the receipt, possession, use, transfer or disposal of radioactive

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9 Stoiber, Carlton; Baer, Alex; Pelzer, Norbert; Tonhauser, Wolfram. IAEA Handbook on Nuclear Law. Vienna: International Atomic Energy Agency, 2003.

10 Number 27 of the matrix is "other"

11 Crail, Peter. "Implementing UN Security Council Resolution 1540: A risk-based approach." *Non-proliferation Review, Vol. 13, No. 2, July 2006.*

material without authorization" , are border controls in the case of illicit nuclear trafficking indeed relevant or a part of nuclear export controls?[12]

The use of export control activities to cover illicit trafficking should keep in mind the objective of such controls, that is, to stop the proliferation of nuclear weapons. Treating border controls for illicit trafficking independently from export controls would signify that illicit trafficking only occurs outside regulatory and legal frameworks, or as the IAEA states, " without authorization." However, illicit trafficking also occurs using false documents and information within these frameworks on the part of proliferators, or even due to the sheer ignorance of the law by exporting companies. Shipping a dual-use good without a license for lack of communication of the law or because of falsification of documents is illicit trafficking that could be curtailed if export controls were more effective.

Even the IAEA, in its stance on illicit trafficking, qualifies its definition by noting that illicit trafficking should " [cover] all unauthorized events involving radioactive materials, irrespective of type and cause, since most of these may only be administrative offenses and matters for the national nuclear or radiological regulatory authority rather than for law enforcement." [13] Yet any such transfer is *always* also a matter for enforcement authorities, because if the national nuclear or radiological regulatory authority fails for whatever reason to effectively curtail an illegal export, border controls are the last step before the export occurs that can stop it. Therefore, regardless of how or why an illegal export can take place, effective border controls can help block dangerous exports before they occur.

As a general definition, border controls are the measures used by countries to monitor or regulate their borders. This is conducted through customs, which control the flow of goods, and the enforcement of controls through border guards or coast guards. Therefore the movement of nuclear materials and equipment must be regulated by a custom agency which can block a potentially forbidden trade flow before it crosses a border. The 1540 Committee matrix identifies border control and technical support of border control activities, which is quite general, but in reality signifies the implementation of many measures. Effective border controls entail the training of personnel, technology and equipment especially regarding radiation detection and border monitoring, and border security, to name just a few components.[14] In case an individual attempts to smuggle radioactive material across a border, authorities must have proper equipment in order to detect such activity during a short period of time. This involves, for example, the use of radiation detectors which sound an alarm if a certain radiation level is surpassed. Radiation detectors can be fixed as portals at border crossings, and can also be used by patrols as hand-held devices to be used in situations where intelligence information has drawn suspicion to specific individuals.[15] In case radioactive material is

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12 "*Prevention of the Inadvertent Movement and Illicit Trafficking of Radioactive Materials.*" IAEA TECDOC Report, Series No. 1311, September 2002.

13 Ibid.

14 Ibid.

15 Bonet Duran, SM; Canibano, J.A; Menossi, S.A; Rodriguez, C.E. "Prevention of the inadvertent movement and illicit trafficking of radioactive and nuclear materials in Argentine border." (paper presented at the 44th Annual Meeting of the Institute for Nuclear Materials Management, 13-17 July, 2003).

found, a system must be in place for radiological emergency response, which involves not just equipment but also the thorough training of personnel. Finally, effective border controls should rely extensively through information sharing with other countries as well as related national agencies handling nuclear exports, such as licensing and enforcement bodies.

The clear difficulty in maintaining effective border controls is the large amount of resources necessary for border control activities. Whether or not a state has nuclear facilities or materials on its territory, border controls are crucial to non-proliferation as they disrupt the flow of illegal transfers, especially in countries which can be used along transit routes.[16] Nevertheless, an effective nuclear export control system cannot rely solely on border controls. This is an issue because of all the assistance made available to states requesting it, border controls tend to be the activities for which the most resources are devoted. The reason behind this is logical: it is easier to measure progress when equipment or training is involved than when the measure is the effectiveness of a law or organization. Assisting counties can send radiation monitors and offer several training programs for customs authorities and consider the mission accomplished. Border controls are absolutely necessary for effective nuclear export controls, but they should never be the sole focus. Other measures should take take place before an export arrives at a border in order to keep illegal trades of nuclear materials and equipment from taking place. Licensing is an integral part of nuclear export controls without which effectiveness in domestic implementation is impossible.

3.3: Licensing

If a domestic export control system is to work effectively, a thorough licensing system should be organized and enforced to control the trade of nuclear materials and equipment. Licensing, in short, refers to permission, whereby a licensee requests permission from a licensor for the freedom to conduct a certain activity.[17] The 1540 Committee matrix lists components of licensing that demonstrate that it is a complex and somewhat confusing process, which not even the most developed countries have succeeded in setting up in an wholly uncomplicated manner. Before examining why it is so difficult to implement adequate licensing operations regarding nuclear exports, it is helpful to examine the components of such operations as listed in the matrix.

The 1540 Committee matrix lists the following activities related to licensing as regards OP 3 (c) and (d) and related matters from OP 6, and OP 10 of Resolution 1540: licensing provisions, individual licensing, general licensing, exceptions from licensing, licensing of deemed exports, national licensing authority, and inter-agency review for licenses. Licensing provisions simply refer to the legal instruments that provide for licensing to take place, such as a law setting up the basis for a domestic licensing system. Such a law should ideally identify the agencies involved in licensing, in which cases licenses are necessary, the procedures involved in receiving a license, and enforcement measures in

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[16] Gabulov, I.A. “Emerging nuclear security issues for transit countries,” in *Radiation Safety Problems in the Caspian Region*. Amsterdam: Kluwer Academic Publishers, 2004.

[17] The etymology of the word license shows that it comes from the Latin *licentia* meaning “freedom, liberty, license.” It is up for philosophical debate, then, why freedom must be granted by a licensor!

case of violations. For example, in the United States, the Nuclear Non-proliferation Act of 1978 spells out in which situations licenses are necessary, while section 57b of the Atomic Energy Act identifies which government agencies are responsible for export control. It is recommended that licensing provisions should be concentrated in one comprehensive law, making it as clear to exporters as possible what steps they must take to receive a license.[18]

A well-working licensing system should account for the difference between individual and general licensing, as specified in the matrix. An individual license is " specific to an individual exporter and covers multiple shipments of specific goods to specified destination(s) and/or, in some cases, specified consignees/end-users." [19] On the other hand, a general license refers to a broad category of exports. Usually in practice exporters determine on a case-by-case basis whether they must apply for a general or an individual license based on the type of good being exported and the destination. A general license usually does not require a specific application but does require a declaration by the exporter if the goods exported exceed a certain value. For an individual license, more paperwork is involved, as well as thorough record-keeping and tracking.[20] In terms of nuclear exports specifically, general licenses usually encompass products containing radioactive materials and nuclear reactor parts in cases where they are exported to certain countries. Other nuclear exports that are either being sent to certain destinations or do not belong in the broad categories listed under general exports require individual licenses. In some cases, licenses are not required at all in order for an export to occur. Exemptions from licensing refers to these situations and by and large covers exports conducted by government bodies and their related contractors and subcontractors.[21] Export control provisions should account for such exemptions by unequivocally stating in which cases exports do not need a license.

One of the trickiest areas of licensing involves the licensing of deemed exports, which the 1540 Committee includes in its matrix. A deemed export occurs when technology is given to a foreign national. In such a case, a trade can take place without crossing borders and still be subject to nuclear export controls. Deemed exports apply to technology or source code, that is, intangible goods, rather than material goods because knowledge can easily be re-transferred to another country while a material good would need a license. In reality, even the export of intangible goods requires control as the section regarding control lists will demonstrate later, but that does not mitigate the necessity of accounting for deemed exports as well.

Implementing controls on deemed exports obviously gives rise to numerous obstacles, first and foremost because such exports are very difficult to track. By definition, two criteria can help determine whether a domestic transfer requires a license. First, a license is required if the person

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[18] Harding, Margaret. "Spaghetti with meatballs: Nuclear export control reform." *The Energy Collective*. October 14, 2010.

[19] Licensing Unit, UK Export Control Organization, August 3, 2009.

[20] Export regulations, customs benefits and tax incentives." United States Commerce Department, Chapter 11.

[21] See US NRC Regulation § 50.11 Exceptions and exemptions from licensing requirements. [40 FR 8788, Mar. 3, 1975, as amended at 65 FR 54950, Sept. 12, 2000]

transferring technology intends to do so to a foreigner. Second, a license is required if transfer of the same technology to the foreigner's home country would require an export license.[22] These controls are especially difficult to implement in a globalized world where information exchange takes place very often among specialists from different countries working in research, academia, and industry.

It would appear that controls on deemed exports inevitably affect some countries more than others, since many states do not have domestic programs related to the nuclear field in which sensitive information could be exchanged within borders. Yet even countries without domestic nuclear programs of any kind must control the activity that takes place not only via borders but also within state territory, as any state with weak controls can be exploited by would-be proliferators. Just as states with feeble border controls can be taken advantage of to ship or trans-ship illegal goods, states with weak or non-existent deemed export laws can be used to transfer information illegally. Controls on deemed exports are therefore necessary to prevent potential future cases as well as account for as much activity as possible as regards the control of nuclear materials, equipment, and technology. Rules on deemed exports should also be well-publicized in fields where exchange of sensitive information takes place.

The last two items on the 1540 matrix related to licensing for exports addresses bureaucratic organization, that is, specifically, setting up a national licensing and an inter-agency review for licenses. This seems like an easy task at first blush-hire staff, train them, create the agency, and voila! In reality, even the states most advanced in their nuclear export control system have a complicated multi-agency tangle of authority, in which different government ministries control the licensing of different nuclear exports. Take the United States, for example. The Atomic Energy Act, in Section 57b, five different government agencies are in charge of nuclear export controls: the Department of Commerce, the Department of Defense, the Department of Energy, the Department of State, and the Nuclear Regulatory Commission.[23] Each of these agencies has different regulations, yet must be in concurrence for a license to be granted.

Necessitating the concurrence of five different government agencies could be seen as a positive sign that exports are scrutinized through five different lenses, thereby decreasing the possibility that a potentially dangerous export could occur. Section 57b, after all, also provides for " an inter-agency coordinating authority to monitor the
processing of [license] requests, predetermined procedures for the expeditious
handling of intra-agency and inter-agency disagreements and appeals to higher authorities, frequent meetings of inter-agency administrative coordinators to review the status of all pending requests, and similar administrative mechanisms." This can be viewed as a model law for countries in which different agencies are in charge of granting licenses, for such inter-agency coordination is listed in the 1540 Committee matrix and also simply a logical necessity when several bureaucracies are in charge of a national and international security issue.

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[22] "The deemed export rule in an era of globalization." United States Department of Commerce, December 20, 2007.

[23] "Assistance for foreign atomic energy activities." United States Atomic Energy Act Section ,57b.

Licensing of nuclear exports can conversely be set up in a more streamlined and efficient way, in which one single agency is entrusted with granting licenses. In a 2010 speech, US Secretary of State Robert Gates admits that the multi-agency system is " not set up to deal effectively with those situations that could do [the US] the most harm in the 21st century" and that " the current arrangement fails at the critical task of preventing harmful exports while facilitating useful ones."[24] While a multi-agency arrangement is ideally supposed to provide a kind of checks-and-balances system in which all interests and demands are met, the incredible " byzantine amalgam," to quote Gate's terminology, ends up being a counter-productive force where mistakes are more likely to occur and exporters can more easily seek out loopholes and circumvention strategies. It is therefore recommended that one single licensing agency is put in charge of all nuclear exports, whether they are munitions or dual-use or deemed or intangible.[25] A single agency would also mean a single database instead of overlapping nuclear export information stored in different locations for use by different agencies. This would not mean that other agencies would never have a say, or that intelligence could not be shared.

While the matrix specifies inter-agency review for licensing, it is strange that there is no measure specified by the 1540 committee regarding information-sharing among different countries regarding licenses. This is especially strange as the Resolution clearly specifies, in OP 7 and OP 8, to " promote dialogue and cooperation on non-proliferation" as well as to take " cooperative action to prevent illicit trafficking." In light of this, the 1540 matrix does not ask whether countries have set up a procedure for informing each other regarding license refusals or other relevant information, such as suspicious end-users or violations. And yet it is crucial for the successful implementation of nuclear export controls, as well as for Resolution 1540 to reach its desired objectives, that licensing officials throughout the world maintain access over sensitive end-users and previous license denials.[26]

Licensing is clearly one of the most important components of an effective nuclear export control system. The 1540 Committee matrix lists the measures states must take in order to achieve a well-working licensing system, but clearly, like with all the other 1540 matrix measures, neither can work in a vacuum. It is not enough to have a licensing system if border controls are weak, for example. All the parts of export controls should be accounted for in order for them to work smoothly. Indeed, an important question remains after discussing licensing: what is to be licensed? That is, which exported goods require such regulation? On the international level, this book has thus far tracked the development of control lists as developed by nuclear export control regimes. On a national level, however, the question of what goods to control is never simple, especially for those countries which are not members of the nuclear export control regimes or who have little experience in managing the trade or transit of such goods. And, as can be expected, the 1540 Committee matrix lists the existence

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[24] Gates, Robert. "Export control reform." (Presented at the Business Executives for National Security, April 20, 2010, Washington DC).

[25] Harding, Margaret. "Spaghetti with meatballs: Nuclear export control reform." *The Energy Collective*. October 14, 2010.

[26] Beck, Michael. "Reforming the multilateral export control regimes." *The Non-proliferation Review*, Vol. 7, No. 2, Summer 2000.

of control lists, and measures related to them. The next section therefore discusses in detail the national implementation of control lists as part of an effective nuclear export control system.

3.4: Control Lists

In order to comply with Resolution 1540, and as part of nuclear export control implementation, countries must enact control lists. A control list is a list of items subject to licenses. Therefore, if an exporter wishes to export a good on the list, they must necessarily be granted permission to do so in order for the export to take place. The 1540 Committee matrix groups several elements of effective control lists together, demonstrating how many factors countries must address in order to comply with the wording of the Resolution. The difficulty lies in identifying what is, after all, an " effective national control list" as stated in OP 6 of the Resolution.

The matrix identifies not just the creation of the list, but the need to update it, to include relevant technologies, to include means of delivery, to establish end-user controls and a catch-all clause, and to account for intangible transfers. All of these components will be dealt with in this section, but it is first necessary to establish a rudimentary understanding of the underlying challenge of adopting a control list. Specifically, the 1540 Committee matrix does not mention exactly how countries should evaluate what items must be included on the control list in order to it to be effective. This is a particularly sensitive question as control lists are often viewed as an obstacle to trade because they " impose restrictions on access to material, equipment and technology for peaceful purposes required by developing countries for their continued development," as stated by members of the Non-aligned Movement[27] Nevertheless, if a country wishes to comply with the Resolution, as it is indeed obligated to do, it must figure out what items to include on the control list. This could be done by examining the control lists of other countries, or of the nuclear export control regimes, or by asking for technical expertise from states in a position to help, the IAEA, or other international organizations.

A typical control list is divided into several parts although this kind of organization varies and therefore it is necessary to embark on a comparative study in order to form an opinion on what could be a best practices model. Taking INFCIRC/254, the list used by the Nuclear Suppliers Group, as an example, the control list organizes different types of materials and equipment according to uses. For example, items for use in gaseous diffusion enrichment are listed separately from those used in aerodynamic enrichment plants. This separation provides information on the type of products associated with potential proliferation activities, especially as regards enrichment activities.

Other control lists vary in structure and do not always group items by uses. China's control list, for example, just has two parts; nuclear materials and nuclear equipment, and non-nuclear materials for reactors. Items are listed under either grouping one after the other, without regard for use, with an explanatory note regarding what the item can be used for in a nuclear context. The EU dual-use

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[27] Final Document of the XIII Conference of Heads of State or Government of the Non-Aligned Movement Kuala Lumpur, 24 - 25 February 2003.
The points mentioned in this document present a serious argument against the Nuclear Suppliers Group and the Zangger Committee.

Regulation 428/2009 assigns a category to materials and equipment depending on what they can be used for in a non-nuclear context, except for Category 0, which lists nuclear material, facilities and equipment. Categories 1-9 cover typical dual-use items that can be used in civilian activities or could potentially also be used in nuclear proliferation, such as special materials, material processing, electronics, computers, telecommunications and "information security", sensors and lasers, navigation and avionics, marine items, and aerospace and propulsion equipment.[28]

After analyzing the control lists of diverse countries, it becomes clear that apart from variations in the organization of items, another significant differentiating factor is the level of specificity of the control lists themselves. In general it should be brought to light here that not all control lists are created equal. The control lists for many countries exist in the form of a half a page or so of general reference to "nuclear materials" or other broad terms, or simply refer to export controls as applying to items listed on "control lists of which are established by international non-proliferation regimes."[29]

It may not seem immediately straightforward why such a general approach falls short of what can be considered an appropriate and effective export control system. After all, if a trained licensing authority is well-informed of the items that should be controlled, perhaps there is no need for specific lists at all. This would be deeply flawed however, for several reasons. First, referring to the control lists of non-proliferation regimes necessitates clearly stating what regimes are referenced. It is more helpful at that point, if a decision is made by a nation to adopt the control list of, say, the Nuclear Suppliers Group, to clearly specify the items on the NSG control list in the national law, and update it regularly. Second, and more importantly, even if the licensing authority knows what items to track, it is much more difficult for exporters to understand what they can and cannot export. An effective export control system should keep a specific and detailed control list to make it easier for exporters to identify what type of license they need to export their goods, and also to understand why their goods may be controlled by explanations in the control list regarding uses and categories.

The logic explaining different control list organization is not directly apparent, nor is it easy to determine which kind of control list is best. It can be inferred from analyzing different ones that a model control list incorporates the following: a list of items for control according to a logical grouping, explanatory notes in order for it to be clear why the stated item is being controlled, and an order which makes identifying items on the list as easy as possible. An Internet-based search system is further recommended in order for exporters to be able to easily look up the controls related to items they wish to export. It should be noted that too much explanation can be counter-productive in terms of publishing sensitive information about nuclear activities, and therefore caution should be used in this matter.

In addition to how control lists are structured and detailed, it is necessary to identify key elements that make them effective. To put it another way, control lists can be judged based not just on

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[28] EU Council Regulation 428/2009, May 5, 2009. It is curious that the list is set up so differently from the INFCIRC/254 list, even though the EU has observer status in both nuclear export control regimes, and almost all EU states are members of them.

[29] This example is taken from the "Law of Georgia on Export Control of Armaments, Military Equipment and Dual-Use Products." Chapter 2, Article IV. Tblisi, April 28, 1998.

their quality, such as how specifically they are written and the clarity of their organization, but also on the quantity of elements that control nuclear exports. Two such crucial elements of control lists, which are mentioned separately in the 1540 Committee matrix, are the inclusion of relevant technologies and means of delivery. It is puzzling that these two measures are listed separately from the general measure of " control list" in the matrix and therefore require closer attention to understand why this is so. In terms of technologies, as phrased by the matrix, the term is probably used to differentiate it from goods and materials. But if technology requires those goods and materials on the list , and if lists usually already state for what technology goods are to be used, why the emphasis on differentiation? Looking at control lists can again help to understand this. Canada's export control list, for example, does not group items differently from technology, but rather defines the term technology in a separate paragraph of its list, paragraph 3-4. Here reference is made to the use of the term in the rest of the document as the " development" , " production" , or " use" of items specified in Group 3, which is a non-proliferation list. Similar ways of treating the term can be found in other control lists, where technology is specified as related to the items on the list. Defining the term in the context of control lists does not however provide much insight into why the 1540 Committee emphasizes the inclusion of technologies in control lists. It can be inferred, therefore, that the Committee specified technologies in order to bring special attention to the fact that identifying materials and equipment is not enough, but must be accompanied by an explanation of the technologies they are used for.

The specification by the matrix for the inclusion of means of delivery follows a slightly different logic. The materials and equipment used for means of delivery are not used specifically in nuclear activities, but contribute to nuclear weapons proliferation by providing the technology and systems that suspected possessors of nuclear weapons have of delivering such weapons. Nuclear weapons can be delivered via ballistic missiles, cruise missiles, or aircraft bombers.[30] These kinds of delivery systems are rather state-biased, however, as they constitute sophisticated systems used by military organizations who care less about cost and more about reliability. A nuclear weapons attack from a terrorist organization most likely would not use such elaborate military technology. According to most scenarios, a terrorist nuclear bomb would produce an uncertain yield of a few kilotons and most likely would be delivered by a simple vehicles such as a truck or a cargo ships.[31] While not accounting for the possibility of a terrorist organization acquiring more advanced technology would be irresponsible, it is therefore important to control delivery systems likewise for that purpose, even if the scenario is nevertheless realistically highly unlikely.

Now that the basic concept of control lists has been identified, it is important to turn to other components, apart from the list itself, that make it more effective in combating nuclear proliferation. A

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[30] Several sub-categories of these three broad means of delivery exist but such a technical discussion is beyond the scope of this book. A fine reference for more on the subject is Palmer, Norman and Norris, Robert. The US Nuclear Arsenal: Nuclear Weapons and their Delivery Systems since 1945. Naval Institute Press, 2009.

[31] J. Carson Mark et al., "Can Terrorists Build Nuclear Weapons?" in Paul Leventhal, and Yonah Alexander, Preventing Nuclear Terrorism. Lexington Books, Lexington, MA: 1987. Also see the Nuclear Threat Initiative's Nuclear Tutorial, Chapter 2.5.

further element of a comprehensive control list, as specified likewise by the 1540 Committee matrix, is the inclusion of end-user controls. These controls are defined as clauses calling for the final recipient of an export to state what they will use the export for. The matrix includes this criteria for OP 6 compliance. An end-user clause must take into account several factors in order for the license to be granted, factors that do not depend solely on the type of export, but also on the recipient. Factors include the reliability of the party involved in the transaction, the ability to separately evaluate every case and the sensitivity level of the end-user, the requirement to separate transactions based on the sensitivity of the product involved, and a differentiation between the various phases of the transaction and the instruments available for end-use control.[32] The inclusion of end-user controls in control lists is an important element of nuclear export controls, especially with regards to dual-use items, because the recipient of an export can shed light on potential proliferation activities. Known suspicious entities on the receiving end of an export can alert authorities of a potential violation of Resolution 1540. In addition, an end-user clause can prevent the re-export of materials and equipment to potential parties seeking to proliferate nuclear weapons, as happened in the case of the A.Q Khan network, for example.

Another element of an "effective" control list includes a catch-all clause. Such a clause is obligatory for members of the Nuclear Suppliers Group, but obviously the 1540 Committee sees is as an important part of 1540 Resolution compliance as demonstrated by the inclusion of such a clause in the matrix. A catch-all clause signifies a clause that can "catch" all types of situations and possibilities, and is commonly included in many types of legal contracts. In the context of nuclear export controls, a catch-all clause aims to regulate exports that could lead to nuclear proliferation, but which are not specifically stated on control lists.[33]

Several models for catch-all clauses exist in various nuclear export control laws. INFCIRC/254/Rev.6/Part 2 of the NSG guidelines in this case represents one of the most clear models for such a clause. The guideline states that "an authorization that shall be required for the export of dual-use items not listed in [Annex I of the NSG control list], if the exporter has been informed by the competent authorities of the Member State in which he is established that the items in question are or may be intended, in their entirety or in part, for use in connection with the development, production, handling, operation, maintenance, storage, detection, identification or dissemination of [WMD]." Article IV of the EU dual-use regulation is another model. The article requires exporters to apply for an export license even if the exported good is not listed in the Annex I control list of the regulation, under two circumstances: First, is the product is in any way intended for use in connection with nuclear weapons; and second, if the item, can be used in a military context in countries under an international arms embargo.

The logical issue that arises after reading the NSG guideline and the EU dual-use regulation is the question of how such intentions can be identified, and by whom. The responsibility of identifying

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[32] Pietsch, Georg. "End-user and end-use controls." (Presented at the 9th International Export Control Conference, Dubrovnik, Croatia, 20-22 October 2008).

[33] Robdrup, Dothe. "*Catch-all.*" Danish Enterprise and Construction Authority, 2010.

this lies with the exporter, at least in the EU context. The Danish export authorities, for example, state that " the exporter himself must seek information about any risks related to his export markets, and the exporter must himself collect information about the end-user and the end-use of the product in the form of an end-user certificate." [34] If national authorities give exporters such a responsibility, it is intuitive that they must supply exporters sufficient information relating to their nation's export control rules, and especially licensing procedures and control lists. However, the exporter, if dubious about any case, can notify their national authority and ask for advice. A catch-all clause, through the joint action of national government and exporter, therefore eliminates the risk of nuclear weapons proliferation occurring due to the omission of a particular item from a control list. It lays responsibility on exporters to verify the reliability of the end-user and the end-use of the product being exported, even if it is not specifically named.

Catch-all clauses are one kind of tool that nations use to block potentially dangerous exports. Along these lines, control over intangible transfers works in a similar manner, as it would be impossible to specifically list and account for every type of transfer bearing a proliferation risk. While the 1540 Committee matrix uses the broad term " intangible transfer," here it is necessary to identify differences between the type of transfer and the way in which it is transmitted, as these are discussed in different ways in literature on the subject. The general term intangible transfer refers basically to knowledge, although ostensibly the transfer of knowledge can be broken down according to oral or manual technical assistance.[35] For example, controlling intangible transfers may include measures to counter the brain drain of nuclear experts who may offer their knowledge and expertise to countries or organizations wishing to build nuclear weapons, or training for employees in the nuclear field regarding the responsibility to keep certain knowledge classified. Controlling not just for knowledge but for the way in which it is transmitted is likewise seeking to control an intangible process. The term intangible technology transfer (ITT) refers to the transfer of software and technology via intangible means, such as fax, e-mail, Internet, or oral transfer.[36]

Accounting for intangible transfers in domestic law can be quite a tricky process. A useful best practices model is provided by a Wassenaar Arrangement document presented at their 2006 Plenary meeting. While the Wassenaar Arrangement controls conventional arms and dual-use goods and technologies, their suggestions for model laws regarding intangible transfers are general enough to be applied in the context of nuclear export controls. Nations implementing intangible transfer controls can therefore use the Wassenaar Arrangement controls as a model. According to the Arrangement, domestic laws should include, according to this model, the following elements: a clear definition of what constitutes an intangible transfer and how such a transfer can occur, specifying in the laws what kinds of intangible transfers are subject to control, and clarifying that controls on such transfers are not

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[34] Ibid.

[35] "Practical aspects of enforcing controls on intangible transfers of technology: German experience." (Presented at the 9th International Export Control Conference, Dubrovnik, Croatia, 20-22 October 2008).

[36] "Strategic Commodities Control System Trade and Industry Department Frequently Asked Questions." Strategic trade control Hong Kong website, http://www.stc.tid.gov.hk/.

applicable to open source information.[37] Effective controls on such transfers likewise necessitate informing industry, academia, and individuals of their responsibility to abide by the law, as well as enforcement by means of penalties, reporting requirements, compliance checks, and surveillance.

In terms of what such a law actually looks like, few examples exist to date because most countries do not have laws in place solely for such transfers. Instead, intangible transfers are usually regulated by different domestic laws regulating diverse activities. In the United States, for example, intangible transfers are regulated by the Arms Export Control Act (AECA), the Export Administration Act, Export Administration Regulation, the Atomic Energy Act, and the Nuclear Non-proliferation Act.[38] Many other countries follow a similar pattern; Hong Kong, for example, admits that intangible transfers do not fall under the Import and Export Ordinance regulating strategic trade controls, but rather are controlled rather indirectly by a Weapons of Mass Destruction Ordinance.

Regulating intangible transfers in this way is certainly better than not accounting for them at all in domestic law, but the ideal would be a specific law addressing such transfers which can be used as an easy reference point. A good example of such a law is Singapore's Strategic Goods Control Act, which creates a permit requirement for the " transmission of controlled strategic goods technology in Singapore by electronic means, or the act of making the controlled strategic goods technology available in Singapore on a computer or server, so that it becomes accessible to a person in a foreign country." [39] These types of transfers subsequently require a permit where the technical specifications and end-use certificate is submitted, and such procedures are likewise clearly stated in the law. Finally, the enforcement of this type of law ideally includes audits of businesses during which information regarding intangible transfers must be made available to audit officers.

Throughout this discussion of control lists, it is quite clear that this area of export controls is quite a tricky affair. In order to meet the " appropriate" and " effective" standard for Resolution 1540 compliance, states must draw up control lists that include a catch-all clause, end-user controls, and controls on intangible transfers, in addition to the items listed that require an export control license. These measures are specified in the 1540 Committee matrix, not necessarily in any particular order, and lead to a discussion of the last and most intuitive component of measures related to control lists: updating them. Updating control lists means keeping abreast of developments and information-sharing in order to keep the lists as inclusive and detailed as possible. This can be done by voluntary information-sharing or assistance programs such as the ones listed under the 1540 assistance program. States can also take existing control lists, as discussed earlier, and check them regularly for updates. For countries that have little or no background in nuclear export controls, it is difficult to imagine that the creation, much less the updating, of control lists can be done without significant outside assistance.

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[37] "Best practices for implementing intangible transfer of technology controls." Drafted at the Wassenaar Arrangement Plenary Meeting, Stokes Australia, December 2006.

[38] "Controls on tangible and intangible transfers of technology." United States Office of Export Control Cooperation, Website: www.exportcontrol.org

[39] Strategic Goods Control Act, Singapore Customs Authority, January 1, 2003.

3.5: Control over the Movement of Exports, Imports and Means

The 1540 Committee matrix lists controls over exports leaving a country, and the ways in which to stop a potentially dangerous export from leaving through the use of border controls, licensing procedures, and control lists. Unfortunately, these mechanisms are not enough in and of themselves, for they several provide loopholes through which potential proliferators can jump. In addition, in case the three categories of national controls- border controls, licensing, and control lists- fail, further measures can be taken that constitute a part of nuclear export controls. These have been designated here in general as control over the movement of exports, imports, and and means. The matrix delineates measures relating to this group as transit control, trans-shipment control, re-export control, control of providing funds, control over transport services, and control over importation. These terms come up frequently in nuclear export control literature, yet the nuances between them, especially as they can sound quite similar, have not been identified, nor linked to how they occur in national nuclear export control legislation.

To begin with, it is useful to group transit and trans-shipment controls together, as they are often mentioned together. It is, however, very difficult to find a definition for such seemingly commonplace terms in the laws referencing them. In fact, analyzing how these terms are used in national export control legislation demonstrates confusion as to how they are used and under what circumstances. In many cases, countries submitting their matrix to the 1540 Committee simply left question marks under these terms. In particular, there seems to be no significant definitional nuance between "transit" and "trans-shipment" as opposed to "re-export" which can be used as an umbrella term for the two.

One possible model that sheds light on these terms is the United Kingdom Export Control Act update, drafted in 2007, that offers supplementary guidance on trade transit and trans-shipment controls. The document defines the difference between transit and trans-shipment very clearly. Transit, according to paragraph 6.1 of the law, refers to a situation in which goods are to be "trans-shipped from one aircraft to another or one ship to another for direct delivery to a non-community country and the goods **do** leave airside or portside environments."[40] Trans-shipment occurs when such goods **do not** leave the stated environments. In both cases, goods pass through a country with a view to re-exportation. From a practical point of view, trans-shipment appears somewhat more difficult to control for the goods being transported do not leave the point at which they were brought into a country on route to their final destination, thus decreasing the probability that they will be detected. In fact, in much of the literature on nuclear trafficking, trans-shipment is referred to many times with little or no mention of transit.[41]

It seems that attention to the details differentiating these definitions is not often given in the nuclear export laws of many countries, leading to what can only be called confusion over the terms.

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40 UK Export Control Act, 2002.

41 "*Nuclear trade outside the Nuclear Suppliers Group.*" Briefing paper prepared by the Australian Safeguards and Non-Proliferation Office, with input from DFAT and other government agencies. January 2009.

For example, The Law of Georgia on Export Control of Armaments, Military and Dual-Use Products defines, in its first article, key terms that occur in the rest of the law. Among these terms, transit is used to refer to the " transfer/movement of products under customs control through the customs territory of Georgia," evidently even when such a transfer or movement is a trans-shipment. Transit in this context then becomes an umbrella term for both scenarios differentiated in the UK act.

The discussion regarding transit and trans-shipment becomes even more curious when looking at re-export. This is a measure that the 1540 Committee matrix uses after transit and trans-shipment measures, yet it is not entirely clear that re-export is a separate measure, rather than an umbrella term. Re-export refers to goods that have been imported into a country and are then exported, that is, the export of imported goods.[42] Re-export necessarily requires transit or trans-shipment; there is nothing mutually exclusive between re-export and either measure. Therefore controlling for transit and trans-shipment automatically controls for re-export. The term re-export, as used in this paper, therefore will be used as a general term encompassing the specific activities of transit and trans-shipment. As far as the use of this term in the 1540 matrix is concerned, it is evident from the amount of blanks and question marks in the country matrices that significant confusion exists over the definition of re-export, especially as in what the matrix seems to identify as a separate measure in addition to transit and trans-shipment.

Clarifying the proper uses of these terms in national nuclear export control law is important because it is necessary for the various entities involved to know what kind of licenses to apply for, and in which cases. Carriers must be aware of their responsibilities when handling goods during trans-shipment and transit, and these responsibilities must be specified clearly in domestic law. These include the responsibility to obtain a valid import license before releasing goods to importers as well as a valid export license before releasing goods to exporters. In addition, carriers must cross-check both licenses for accuracy. From the side of national governments, a balance must be struck between controlling for potentially illegal trade while not hurting legitimate business transactions.

Re-export control measures, as used here as an umbrella term for transit and trans-shipment controls, are extremely important to an effective nuclear export control system because they constitute a further step in the effort to preclude nuclear proliferation by what is referred to as the "no undercutting principle."[43] In the majority of nuclear trafficking cases, countries which have weak nuclear export controls are used as re-export hubs. If export control laws are too tough in the countries from which goods are being exported and imported, a third country with weak controls will be sought out in order to make the illegal transaction possible. Therefore strong re-export controls prevent the "undercutting" of the control systems of a country's trading partners, thereby precluding the opportunity to exploit weak controls.

The 1540 Committee matrix cushions nuclear export controls with measures regulating the means by which proliferation can take place, such as funds and transport services. Domestic law must

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42 Financial Times Lexicon, http://lexicon.ft.com/

43 Lau, Vivian. "Trans-shipment and transit controls on strategic commodities in Hong Kong." (Presentation to the 9th International Export Control Conference, Cavtat-Dubrovnik, Croatia, 20-22 October 2008).

account for the provision of these two means to undercut efforts by entities seeking to engage in illegal trade. Such provisions must be underscored by intelligence-sharing and strong enforcement in order to identify situations in which funds and transport services should not be made available. The 1540 Committee further identifies control over importation as an export control measure, which is quite vague, but buoys border control, licensing, and re-export control measures.

3.6: Extraterritorial Applicability

The final matrix measure identified by the Committee relating to OP 3 (c) and (d) and related matters from OP 6, and OP 10 of Resolution 1540 is extraterritorial applicability. This is an important legal measure establishing the ability of a government to exercise authority beyond its typical boundaries, in this case prosecuting individuals for extraterritorial violations of Resolution 1540.

Extraterritorial jurisdiction has become one of the most widely discussed issues regarding national implementation of the Resolution and deserves a detailed analysis here. The ability of states to assert criminal jurisdiction is based on five generally accepted principles: territoriality, when acts occur within a country; the nationality principle, where a state asserts jurisdiction over its citizens regardless of where a crime has been committed; the passive personality principle, where a state exercises jurisdiction based on the victim of a crime being a citizen regardless of where the crime occurred; the effects principle, where the crime has a significant effect on a state's territory and interests even if it occurs extra-territorially; and universal jurisdiction, where jurisdiction applies to crimes that are universally condemned.[44]

Establishing extraterritorial applicability in domestic law relating to nuclear export controls is important for several reasons. First, it inhibits the ability of individuals engaged in nuclear weapons proliferation to operate in or seek refuge in a state that will not prosecute them. Second, it broadens the ability of states to prosecute acts of nuclear proliferation, thereby helping achieve the goals of Resolution 1540. This means expanding the ability of states with stronger nuclear export control laws to hold to account actions committed in states with weaker laws. Third, extraterritorial applicability strengthens the norm against nuclear weapons proliferation as an international crime that all countries should work together to prosecute.

While implementing extraterritorial applicability measures in domestic law appears to be an effective and logical way to strengthen the enforcement aspect of nuclear export controls, it is perhaps one of the provisions most weakly implemented by states as exemplified by their 1540 reports. The reasons for this will be assessed in the next chapters, where compliance with the Resolution will be examined and analyzed.

3.7: Information

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[44] Gibson, Jennifer and Shirazyan, Sarah. "*Legal cooperation to control non-state nuclear proliferation: Extra-territorial jurisdiction and UN Resolutions 1540 and 1373.*" Paper for the Conference on Cooperation to Control Non-State Nuclear Proliferation: Extra-Territorial Jurisdiction and UN Resolutions 1540 and 1373, Washington DC, April 4-5, 2011.

The 1540 matrix groups measures necessary for the appropriate and effective implementation of Resolution 1540 by their relevance to different operative paragraphs of the law. For this reason, the last part of the matrix specifies seven measures related to OP 6, 7, and 8(d), and in this vein groups control lists, assistance, and information, all together, and moreover, does not specify a difference between such measures as related to biological, chemical, or nuclear weapons proliferation. This is puzzling as not only are the inclusion of control lists here duplicating from previous parts of the matrix, but it somehow undercuts the importance of all three of these types of measures to group them together for all three groups that could be used in WMD. Furthermore, seeing as how assistance is often offered or requested for specifically one type of activity, asking countries to fill out the matrix without more specificity in this last part diminishes the accuracy and effectiveness of the matrix results.

Be that as it may, the measures that are of interest to nuclear export controls, and in fact should probably be grouped with the other nuclear export control measures, is information sharing with the public and industry. OP 6 (d) of the Resolution indeed calls upon states to "develop appropriate ways to work with and inform industry and the public regarding their obligations under such laws." There is no reason, therefore, to group this measure with assistance and control lists rather than with specific nuclear export control measures from the previous group of matrix activities related to OP 3 (c) and (d) and related matters from OP 6, and OP 10. Information sharing will therefore be treated here as if it were part of the previous group of measures because it is absolutely crucial to a comprehensive and effective nuclear export control system. The data used in next chapter's country analysis of national implementation of nuclear export controls will likewise use the measures "information for public" and "information for industry."

Informing the public and industry is a difficult and intensive task requiring cooperation and coordination by the national authorities charged with its undertaking. As far as industry is concerned, national governments have an obligation to work closely to, inform them of their obligations under national nuclear export control law. Because industry lies closest to users of materials, equipment, and technology that could engage in illicit diversion throughout the supply chain, keeping strong lines of communication between the government and industry increases the probability that such illicit activity will be detected and reported.[45] In addition, if the law as well as examples of enforcement are effectively communicated to industry, a culture of self-regulation will develop whereby companies refrain from engaging in possibly illegal activity due to damage not only to their company if caught, but to the ripple effect such an action may have on their entire industry.

Informing industry can take on many forms. The use of documents, such as pamphlets and books providing information regarding the law, can be handed out to parts of industry that should be aware of nuclear export control law. Agency representatives should keep contacts within these parts of industry in order to keep lines of communication open. Finally, laws and policies must be made

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[45] Hund, G and Seward, A. "*Broadening industry governance to include non-proliferation*." Pacific Northwest Center for Global Security Report." November 11, 2008.

easily accessible-that is, members of industry should not only know where to find such information, but once found, it should be clear, organized, and helpful.

A similar logic applies for informing the public. While it is unrealistic to expect the general public to have a proficient and detailed understanding of the details of nuclear export controls, even a rudimentary understanding that such controls exist and work towards keeping the world safe is not. Informing the public to some extent also helps stave off the probability that ignorance of the law will be used as a retroactive excuse to export something that should not be exported. In the end, it is perhaps intuitive that all the national legislation and enforcement in the world won't work effectively against nuclear weapons proliferation if the public and industry is not properly informed of their duties and responsibilities. Especially due to the complexity of licensing regarding nuclear materials and equipment, the public and industry must understand the objectives of nuclear export controls as well as what procedures to follow.[46]

4: Conclusion on National Implementation and Compliance

As of March 2011, 26 states still have not submitted any documentation to the 1540 Committee regarding their compliance with the Resolution.[47] Of those countries that have submitted reports, almost none have implemented all of the measures identified by the 1540 Committee matrices. It is therefore an initial starting point to mention that submitting a report to the 1540 Committee is not equivalent to fully complying with Resolution 1540.

It is further important to once again reiterate why this paper about nuclear export controls has focused so keenly on Resolution 1540. Considering the importance of nuclear export controls in combating nuclear weapons proliferation, UN Resolution 1540 remains the strongest international instrument requiring countries to take action in this regard. The Nuclear Non-proliferation Treaty does require states to broadly combat nuclear weapons proliferation through export controls, but it does so without the kind of support backing Resolution 1540, especially in terms of the specific measures necessary for the effective implementation of nuclear export controls. Using the reporting system required by the 1540 Committee, as well as the individual 1540 matrices, it is possible to track state progress in detail. It is not enough to have a check box for " nuclear export controls" in order to determine whether a country has them or not. Instead, each part of nuclear export controls must be implemented for this non-proliferation strategy to work. Each part is like a component of a complicated machine; if one piece does not exist or does not work, the entire machine risks malfunction.

The objective of this paper has been to develop a best practices model in order to track and analyze progress regarding the national implementation of nuclear export controls, as well as to have

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46 Beck, Michael. "Reforming the multilateral export control regimes." *The Non-proliferation Review*, Vol. 7, No. 2, Summer 2000.

47 (Cape Verde, Central African Republic, Chad, Comoros, Congo (Republic of), Democratic People's Republic of Korea, Equatorial Guinea, Ethiopia, Gambia, Guinea, Guinea Bissau, Haiti, Lesotho, Liberia, Malawi, Mali, Mauritania,Mozambique, Rwanda, Sao Tome and Principe, Solomon Islands, Somalia, Swaziland, Timor-Leste, Zambia, Zimbabwe)

a rubric from which to measure shortcomings and obstacles. Developing this model has necessitated breaking down specific nuclear export control measures individually and analyzing in what way such measures can be implemented for the best result. A best practices model, after all, seeks to find ways of reaching objectives in the most efficient and effective manner. This paper has made an attempt at developing such a model, however the logical next step is aiding countries to achieve in their national implementation the closest possible proximity to the model.

The Big Table
An information tool on items listed for export controls

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Abstract:

Global trade data are widely collected and made available by national statistical services and dedicated data providers. JRC has carried out a support task to IAEA to identify main trade data sources and explore uses for safeguards. In this context a software tool has been developed in support to trade analysis. The tool, called The Big Table (TBT), allows IAEA's analysts to search control lists, identify items of interest, and relate these to technical documentation and descriptors needed to retrieve relevant trade data.
A distinguishing trait of TBT is that it enables searching in an 'inter-related way' a collection of reference documents for export controls on nuclear and nuclear-related items. Reference documents include: regulatory documents, technical handbooks and the Harmonized System, i.e. the taxonomy of goods by the World Customs Organization used by exporters to declare trade to customs. By 'inter-related' search it is meant that, in TBT, semantically related items listed in different documents of the collection are put in correspondence by ad hoc correspondence tables. In this way, any document can be taken as starting point of a search, and retrieve complementary perspectives on items of interest given by other documents in the collection. As such TBT can serve a variety of tasks and communities underpinning export controls, including the rating of items by licensing authorities and commodity identification by customs officers.

Keywords: Export controls; trade analysis; Harmonized System; licensing; customs; information tool.

1. Introduction

The Big Table (TBT) [1] is a software tool designed to:

- Browse and search a collection of *reference documents* listing and describing items relevant to IAEA safeguards, export controls and case studies on nuclear-related trade.
- By the above process, select items of interest to specific *case studies related to nuclear trade*.
- Map selected items to *Harmonized System* codes (HS) [2]. HS is the taxonomy of goods developed by the World Customs Organization (WCO) used by exporters and importers to declare commodities to customs. These declarations are at the basis of *global trade data* published in *web data services* [3]. HS codes are instrumental to the retrieval of trade data records.
- Export HS codes in formats suitable to query web trade data services and retrieve records about trade of interest for analysis.

In short, TBT offers functionalities for analysts to perform steps preparatory to the retrieval and analysis of data records pertinent to case studies on nuclear trade. TBT does not provide access to

trade data services, nor tools to analyze data records. Data retrieval and analysis are performed outside TBT[1].

This paper is structured as follows. Section 2 introduces web data services on global trade. For an extended review, the reader is referred to [3]. Section 3 sketches how global trade data can be used in the context of trade-related case studies. An illustrative case on the trade of export-controlled machine tools is presented. Section 4 presents a collection of reference documents related to International Atomic Energy Agency (IAEA) safeguards and export controls consulted by trade analysts. These include trade regulatory documents, technical handbooks and the Harmonized System itself. The concept of *correspondence tables* between references documents is introduced in Section 5. Correspondence tables identify related items appearing in different reference documents. *Correspondence tables are at the core of TBT*: they create a complete view on items, covering regulatory aspects from several perspectives (i.e. IAEA's, export licensing, customs controls) as well as items' technical descriptions. In view of trade analysis, correspondence tables provide HS descriptors for these items to enable the retrieval of trade data from web data services. Section 6 sketches TBT's workflow. Section 7 provides a comparison between TBT and related information tools developed in the context of export controls. The paper concludes by outlining ongoing and future developments on TBT and pointing to new application opportunities in the area of export controls.

2. Web data services on global trade

Data services on global trade [3] referred to in this paper are *open source*. The data are available to anyone either for free or by a subscription fee. The information provided by trade data services has a regulatory origin as it stems from declarations made by traders to *national customs authorities*. Customs data are collected at national level and, by decision of individual States, published in *transactional* or *statistical* formats.

Transactional data are largely equivalent to declarations made by importers/exporters to customs. Declared data fields subject to disclosure may include:

- A code classifying the commodity traded; e.g., according to the HS product nomenclature;
- Free text description of the commodity;
- Quantity, expressed in weight or number of items;
- Value;
- Date of shipment;
- Country/port of import/export;
- Party names.

Statistical data on trade are derived by aggregating transactions data by country, trade flow (import or export), reference period of time (months, years) and product categories as specified by the HS. As an example, COMTRADE [4], by the United Nations Organisation, provides annual series of trade data for 150 reporting countries. COMEXT [5], by the Statistical Office of the European Union (EU), is a second example. Focused on European reporting countries, COMEXT provides monthly records on EU trade.

The practical use of global trade data is straightforward except for the specification of HS codes needed to retrieve the data. This requires not only a good understanding of how the HS goods' taxonomy is organized, but also knowledge of customs procedures and statistical practices in data processing which can be specific to each country, commodity, trade flow or range of values. This is illustrated in the next Section by means of an example.

[1] TBT includes, though, a web browser to connect to subscribed trade data services.

3. Addressing trade case studies: general steps and one example

Global trade data can be used in the conduct of trade case studies. A case study may start with a piece of information or a hypothesis about trade activities between entities (e.g., countries, companies) on items subject to safeguards, export controls, or other proliferation-sensitive items.
The goal of a case study is to consult web data sources on global trade to:

- Confirm or deny the information/hypothesis;
- Find new, related information.

Example. *A case of trade in high-precision machine tools with unclear end-use has been reported by the press. Articles report that several high-precision machine tools have been exported from Country A to Country B over a period of a few month. Is the alleged trade confirmed by global trade data sources?*

Before consulting web trade data services, a preliminary step is to identify clearly items of interest to the case study at hand. This is supported by expertise on items' and a set of *reference documents* (Section 4) where materials and equipment subject to export controls are listed, defined and described.

Example. *Machine tools subject to export controls are listed in the Nuclear Suppliers Group 'Guidelines for Transfers of Nuclear-Related Dual-Use Equipment, Materials, Software and Related Technology' [6]. Machine tools of interest here include turning machines (item 1.B.2.a. in [6]), milling (1.B.2.b.) and grinding machines (1.B.2.c.) for removing and cutting metals, ceramics or composites.*

With items of interest identified, it is necessary to *map these items to HS descriptors* in order to retrieve relevant trade data[2]. Mapping items of interest to nuclear-related trade to HS is not trivial, because:

- HS codes only approximate targeted items.
- Exporters may (intentionally or unintentionally) declare trade to customs authorities in HS categories aside from the correct ones.

Trade analysts must determine, on a case-by-case basis, which HS codes to use to best address the case study at hand. Factors to take into account include: the accuracy of HS codes with respect to the targeted items, the expected volume of trade for reporting countries, the likelihood of intentional or unintentional incorrect declarations. A starting point for the selection of HS codes is given by existing *correspondence tables*. Developed by experts of the HS, these tables map items listed for export controls to HS codes. A table we make reference to is the EU 'Correlation Table' [7] [8] (CT) developed and maintained by the European Commission, DG TAXUD: CT maps to HS items listed for export controls [9] by EU Member States.

Example. *Export controlled machine tools (turning, milling, grinding machines) are associated to a number of HS codes by the EU Correlation Table. These are reported in Table 1. None of these codes is specific to export-controlled machine tools. For example, no precision-related parameter appears in the HS explanatory notes. Indeed HS is a taxonomy designed to describe generic categories and the most common ones from the trade point of view. This limitation implies that trade data retrieved by HS codes need then to be analysed by trade-related criteria that can, in favourable cases, reduce the inherent data ambiguity. See follow-up point in the Example.*

[2] By definition, trade data referred to in this article do not generally include trade undeclared to customs. Some countries include such trade when discovered.

HS Code	*HS Explanatory note*
8457.10.	*Machining centres for working metal.*
8457.20.	Unit construction machines "single station", for working metal.
8457.30.	Multi-station transfer machines for working metal.
8458.11.	*Horizontal lathes, incl. turning centres, for removing metal, numerically controlled.*
8458.91.	Lathes, incl. turning centres, for removing metal, numerically controlled (excl. horizontal lathes).
8459.10.	Way-type unit head machines for drilling, boring, milling, threading or tapping metal.
8459.21.	Drilling machines for working metal, numerically controlled (excl. way-type unit head machines).
8459.31.	*Boring-milling machines for metals, numerically controlled (excl. way-type unit head machines).*
8459.51.	Milling machines for metals, knee-type, numerically controlled.
8459.61.	*Milling machines for metals, numerically controlled (excl. way-type unit head machines, boring-milling machines, knee-type milling machines and gear cutting machines).*
8460.11.	Flat-surface grinding machines, for working metal, in which the positioning in any one axis can be set up to an accuracy of at least 0,01 mm, numerically controlled.
8460.21.	Grinding machines, for working metal, in which the positioning in any one axis can be set up to an accuracy of at least 0,01 mm, numerically controlled (excl. flat-surface grinding machines and gear cutting, gear grinding and gear finishing machines).
8464.20.	Grinding or polishing machines, for working stone, ceramics, concrete, asbestos-cement or like mineral materials or for cold-working glass (excl. machines for working in the hand).
…	…

Table 1 – Possible HS codes for declaring trade of machine tools to customs.

With HS codes selected, a plan of queries on trade data services is designed taking into account the questions addressed by the case study and the range of services available on trade data. Parameters that come into play are: the geographical coverage of the case study, the time frame addressed, the type and granularity of the data required (i.e., transactional or statistical, as defined in Section 2), and the cost (if any) of accessing the data. For example, querying for statistical data requires the specification of the following dimensions:

- A reporting country;
- HS codes related to the items of interest;
- A trade flow (import, export);
- A time period.

Example. *We query trade data services for reporting Country A exports to Country B. HS codes for queries are listed in Table 1. The time period is a few consecutive months in a given year.*

Queries return the list of partner countries for which trade on the query dimensions exist, specifying *value* and *quantity* of the trade over time. Results of queries are presented in tabular form for analysis and interpretation. The criteria adopted for the analysis are, in general, specific to the commodities at hand. Yet one informative *generic* criterion is the 'trade unit value', defined as the ratio between value and quantity traded for a given HS, in a time frame (e.g. month or year), between traders (countries in the case of statistical data or companies in the case of transactional data). For limited quantities traded, the trade unit value approximates the declared price at which commodities have been traded. Knowing the market price for items targeted in a case study, one can compare it with the trade unit values of retrieved data records. Data points close to market price of targeted items are highlighted as points of interest. These may confirm alleged information or provide new insights in the case study.

Example. *Table 2 and Table 3 show value and quantity of exports declared by Country A to B on HS codes of interest in the time frame of the case study (M, …, M+4). The data could refer to the alleged trade of machine tools in that the trade unit value (Table 4) is compatible with market prices for export-controlled machine tools.*

HS Code	Unit	HS Explanatory note	Value in United States Dollars				
			M	M+1	M+2	M+3	M+4
845710	USD	Machining Centers For Working Metal	0	1076148	0	0	0
845811	USD	Horizontal Lathes For Removng Met Numrcal Controld	870106	0	0	0	0
845931	USD	Boring-Milling Mach Remove Met Numerical Controlld	757020	1868760	0	0	0
845961	USD	Milling Mach N Knee Type Remov Met Numerical Contl	0	2815177	0	0	2737801

Table 2 – Value (USD) of Country A exports to Country B.

HS Code	Unit	HS Explanatory note	Quantity				
			M	M+1	M+2	M+3	M+4
845710	NO	Machining Centers For Working Metal	0	2	0	0	0
845811	NO	Horizontal Lathes For Removng Met Numrcal Controld	2	0	0	0	0
845931	NO	Boring-Milling Mach Remove Met Numerical Controlld	2	2	0	0	0
845961	NO	Milling Mach N Knee Type Remov Met Numerical Contl	0	1	0	0	1

Table 3 – Quantity (number of items) of Country A exports to Country B.

HS Code	Unit	HS Explanatory note	Unit Value (United States Dollars)				
			M	M+1	M+2	M+3	M+4
845710	USD	Machining Centers For Working Metal	0	538074	0	0	0
845811	USD	Horizontal Lathes For Removng Met Numrcal Controld	435053	0	0	0	0
845931	USD	Boring-Milling Mach Remove Met Numerical Controlld	378510	934380	0	0	0
845961	USD	Milling Mach N Knee Type Remov Met Numerical Contl	0	2815177	0	0	2737801

Table 4 – Unit value (USD) of Country A exports to Country B.

Several of the methodological steps presented in this Section are supported by TBT. This will be outlined in Section 6. The following Sections 4 and 5, expand on the reference documents and correspondence tables used in TBT.

4. Reference documents

The identification of items relevant to trade case studies requires the consultation of a collection of reference documents where export-controlled items are listed and described. The collection of documents includes *regulatory documents* and *technical handbooks* on controlled items, as well as the Harmonized System. TBT includes the reference documents listed in Table 5 hereby briefly introduced. While all regulatory documents are public, some handbooks are for IAEA use, and others have been developed for States Members to the Nuclear Suppliers Group (NSG), as specified in Table 5. To respect the document access rights, the TBT distribution bundle includes only the public

access documents i.e. the regulatory documents and the Harmonized System. Technical handbooks must be available at the installation site to become visible in TBT.

Document	*Access*	*Color*
INFCIRC 540c Annex II (1997)	Public	
INFCIRC 540 Annex II HB (2004)	IAEA	
INFCIRC 254 1 rev9 (2007)	Public	
INFCIRC 254 1 HB (2002)	NSG	
INFCIRC 254 2 rev7 (2006)	Public	
INFCIRC 254 2 HB (2003)	NSG	
GRL Annex III (2002)	Public	
GRL Annex III HB (1998)	IAEA	
EC Reg 428/2009 Annex I (2009)	Public	
EC Reg 423/2007 Annexes I, II (2007)	Public	
Harmonized System (2002)	Public	

Table 5 – TBT's collection of reference documents.

- **INFCIRC 540c Annex II of the Additional Protocol** (AP) [10] lists especially designed or prepared equipment (single-use) and non-nuclear material for the reporting to the IAEA of exports under AP[3]. This list of items is based on the 1995 version of the 'Trigger List' set by the Zangger Committee and adopted by the NSG (see next paragragh). **INFCIRC 540c Annex II HB** [11] is a handbook developed for IAEA on items in AP Annex II. It expands the definitions given in [10], describes the items' appearance as manufactured or packaged, and lists the HS codes applicable to the items.
- The IAEA publishes as **INFCIRC 254 1** the NSG 'Guidelines for the Export of Nuclear Material, Equipment and Technology' [12] (referred to as NSG Part 1 or the Trigger List) and as **INFCIRC 254 2** the NSG 'Guidelines for Transfers of Nuclear-related Dual-use Equipment, Materials, Software and Related Technology' [6] (NSG Part 2). By 'dual-use' items it is meant items that have nuclear and non-nuclear applications, as defined by the NSG. Both NSG lists are updated regularly. **INFCIRC 254 1 HB** [13] and **INFCIRC 254 2 HB** [14] are the technical handbooks developed for NSG members for these lists.
- In 1991 Annex 3 of the IAEA's Ongoing Monitoring and Verification established a list of nuclear and nuclear-related items whose export was either prohibited to Iraq or subject to controls. The list, referred as Iraq Goods Review List (GRL) or **GRL Annex III** [15], includes both single-use items (i.e., especially designed or prepared for the processing, use or production of special fissionable material) as well as dual-use items. **GRL Annex III HB** [16] is the associated handbook: items' definitions are expanded by an introductory note, a description of the typical appearance of the item, health and safety cautions associated with it, typical nuclear uses, and site-specific photos taken by IAEA inspection teams in Iraq.
- The European Union **EC Reg 428/2009 Annex I** [9] implements internationally agreed dual-use controls including the Wassenaar Arrangement, the Missile Technology Control Regime,

[3] Article 2.a.(iv).

the Nuclear Suppliers' Group, the Australia Group and the Chemical Weapons Convention. This document provides a single list of items and technology drawn from export controls lists, including the NSG Guidelines, Part 1 and Part 2.

- Besides general export controls lists, it is relevant to consult lists that restrict the export of specific items to particular destinations to implement, for example, resolutions adopted by the United Nations Security Council. This is the case with **EC Reg 423/2007** [17], the European Council Regulation concerning restrictive measures against Iran. In addition to EC Reg 428/2009 Annex I, Regulation 423/2007 (and amendments) lists items whose export to Iran is either prohibited (Annex I) or requires authorization (Annex II).
- Trade data available from web services are retrieved by Harmonized System [2] codes. HS is the reference taxonomy for commodities adopted by customs adhering to WCO, trade associations and statistical offices in the majority of countries. HS is based on about 5,000 commodity groups organized within 22 Sections in a hierarchy made up of: Chapters, Headings, and Subheadings. Each level in the hierarchy is identified by a HS code and an explanatory note. Codes are 2-digit for Chapters, 4-digit for Headings and 6-digit for Subheadings.

Figure 1 sketches the relationship between documents in terms of *types of items* listed in each document. Rectangles represent sets of items referred to by documents. Across the document collection, these can be identical, disjoint or intersecting, or included.

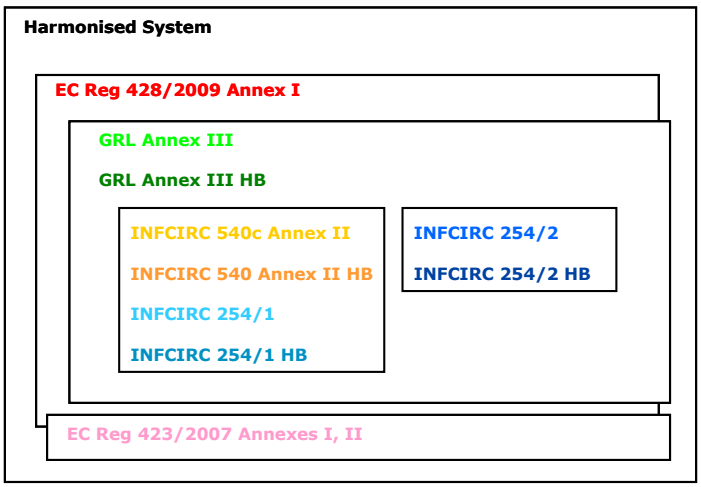

Figure 1 – Approximate relationship between reference documents in terms of items coverage.

5. Correspondence tables between reference documents

TBT's document collection describes items from different perspectives, and with different levels of information. Regulatory documents 'define' controlled items. Handbooks provide information on the items' manufacturing process, appearance and uses. The HS provides indications of customs categories that more likely are used by exporters of listed items. It is then relevant for a trade analyst to search, read and use documents in a cross-referenced way to get a complete picture on items. This was the drive to develop TBT. TBT's primary purpose is to identify, present and manage correspondences between items described in the collection of reference documents. It also helps to identify HS descriptors relevant to a particular item in view of retrieving trade data records from web data services.

Trade analysts can search items in the document collection either by text or by *correspondence tables*. In general terms, correspondence tables relate items listed in different documents of the collection. Some tables identify equivalent items appearing in different reference documents. Other correspondences associate items with HS codes for use in trade data retrieval and analysis.

In TBT correspondence tables are either *native* or *derived*. Native correspondences are made by domain experts by comparing pairs of documents. For example, the EU 'Correlation Table' [7] between EC Reg 428/2009 Annex I and HS is native. Derived correspondences can obtained by

composing existing correspondences (whether native or derived). Given three documents D_1, D_2, D_3 and two correspondences $D_1 \rightarrow D_2$, and $D_2 \rightarrow D_3$, a third correspondence is derived between D_1 and D_3 by: $D_1 \rightarrow D_3 = D_1 \rightarrow D_2 \rightarrow D_3$. This is useful in different ways. *First*, and in view of trade analysis, it is of interest to make relevant HS codes available to items *whatever* the reference document they appear in. For example, the HS codes associated by the Correlation Table to nuclear-related items in EC Reg 428/2009 Annex I can be 'inherited' by the same items listed in INFCIRC 254, 1 and 2. *Second*, the composition operator facilitates the incorporation of revisions of regulatory documents that are already part of the collection. It suffices to define in TBT the correspondence between the new and old version of the document. This is rapidly done using the comparison tables that accompany the revisions of regulatory documents. Figure 2, left, shows as solid lines native correspondences between pairs of documents. The European CT is indicated, relating EC Reg 428/2009 Annex I to the HS. In Figure 2, right, dashed lines show derived correspondences connecting INFCIRC 254, 1 and 2, to the HS. These are obtained by composing native correspondences: the ones connecting INFCIRC 254, 1 and 2, to EC Reg 428/2009 Annex I, and the CT [8]. We have derived and embedded in TBT all meaningful correspondences between documents in the collection for these to be immediately available to the analysts.

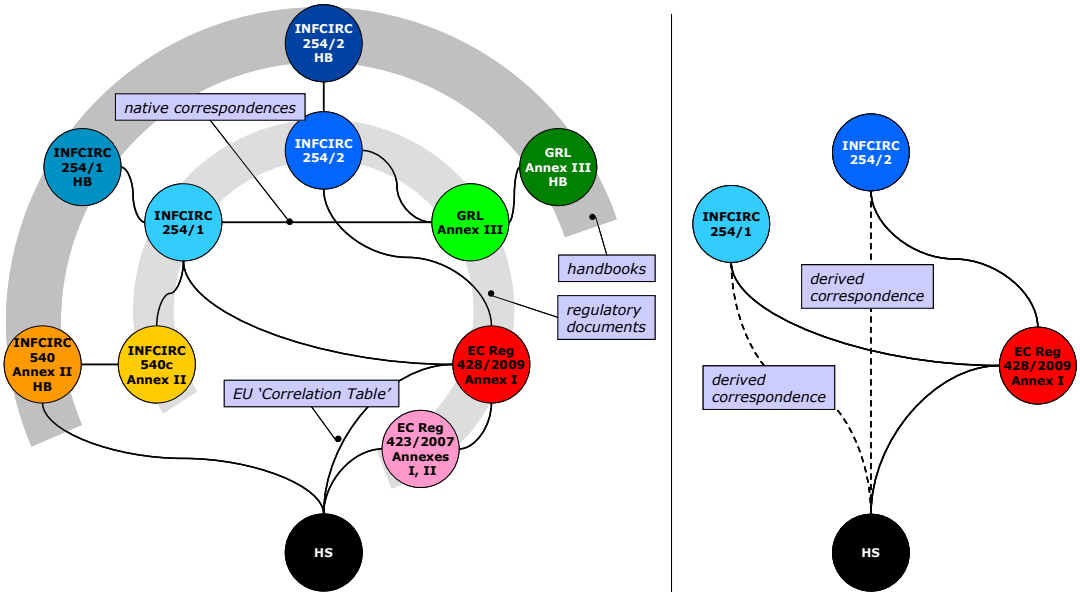

Figure 2 – Left: Native correspondences between reference documents. Right: Correspondences derived (dashed lines) by composing native correspondences (solid lines).

A strong point in TBT is that any document in the collection can be taken as a focus document and retrieve complementary information from the other documents through correspondence tables. This feature enables serving different analyst profiles.

For example, Figure 3, left, captures the perspective of EU licensing authorities for export-controlled items. For the rating of export license requests, the focus reference document is EC Reg 428/2009 Annex I. Analysts need then to access handbook information on items under licensing to support the rating process. For nuclear and nuclear-related items, the handbooks on INFCIRC 254 1 and 2 are relevant. In TBT these can be accessed directly through correspondences attached to EC Reg 428/2009 Annex I items. In a sense, these correspondences create a *virtual handbook* on a number of items listed in EC Reg 428/2009 Annex I.

Figure 3, right, iIllustrates another perspective, this time related to EU customs controls. In this case the focus document is the Harmonized System, because HS descriptors appear in declarations of goods to be exported. Through HS descriptors and the CT correspondence table customs officers get an indication on whether goods under export *could* be listed under EC Reg 428/2009 Annex I. Further, customs can access handbook information on these items and get descriptions of their typical appearance, and health and safety cautions associated with them.

These few examples show how TBT could support several information and decision processes related to export controls and commodity identification of 'listed items'.

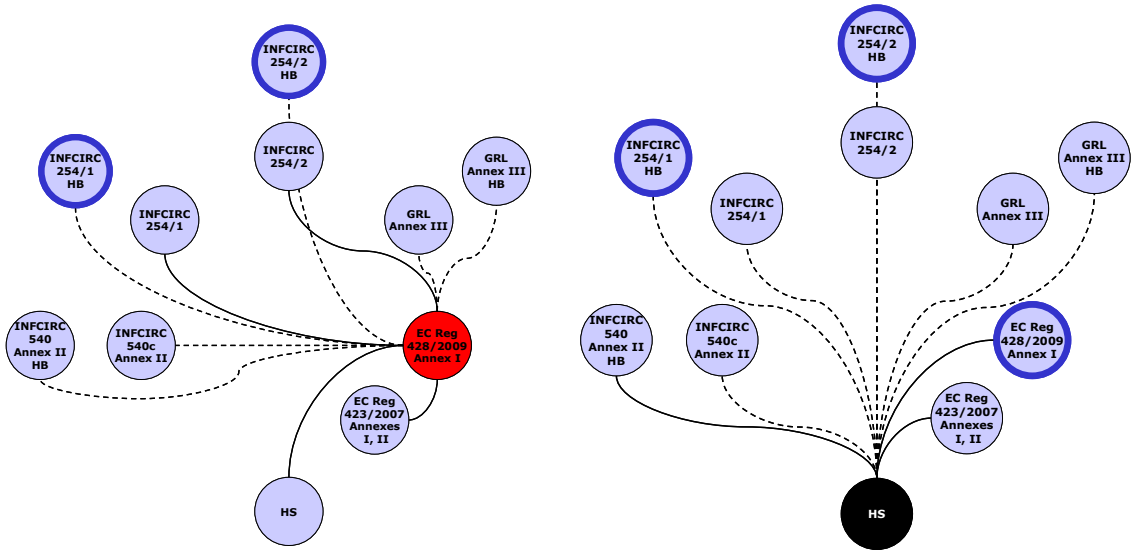

Figure 3 – Left: Correspondences (native and derived) from EC Reg 428/2009 Annex I to other documents in the collection. Right: Correspondences (native and derived) from Harmonized System to other documents in the collection.

6. TBT's work-flow

This Section illustrates TBT's work-flow (Figure 4).

TBT can be seen in first place as an information tool on export-controlled items. These items are defined and described in a collection of reference documents including regulatory documents, technical handbooks and the Harmonized System (Table 5). Documents appear in TBT as *lists of items* and as PDF files. The list of items format is stored in a database. Documents are tables in the database with fields *Code, Title, Description, Note, User Note* for each listed item.

TBT provides functionalities to search the document collection by text and by correspondence tables. Text search can be run on a single document or on multiple documents. Text search can be restricted to selected fields (*Code, Title, Description, Note, User Note*). Returned records can be read as structured text or in the original PDF documents.
Correspondence tables identify related items appearing in the document collection. For example, correspondence tables connect items appearing in regulatory documents to their technical handbook equivalents –if any. Likewise they identify Harmonized System descriptors for items listed in regulatory documents.

Items of interest (for example, to a trade-related case study) can be selected for further processing. Typically items are copied to:

- *Excel* tables – For documentation purposes, it is useful to create tables of items that have been retained as relevant to a given case study. These may include HS descriptors.
- The *Clipboard* – In view of querying web data services on global trade, selected Harmonized System codes need to be formatted in a way compatible to a data service's query mask. This is done in the *Clipboard* by the *Export Codes* tab.

Web data services are queried 'outside' TBT. To this goal, TBT web browser can be used. It stores as Favorites the web addresses of subscribed data services.
Red arrows in Figure 4 depict the main TBT work-flow starting from document search, to items selection, HS codes export and trade data queries. Figure 5 shows the search & view interface to search TBT's document collection.

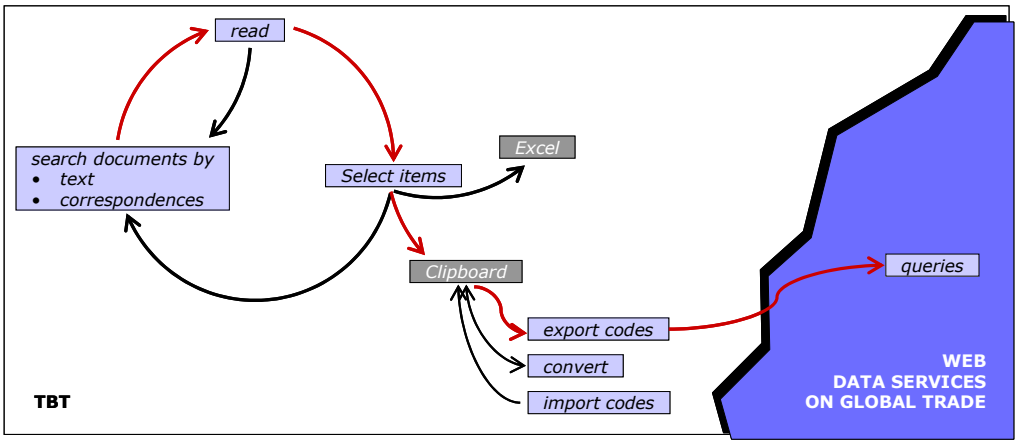

Figure 4 – TBT's work-flow.

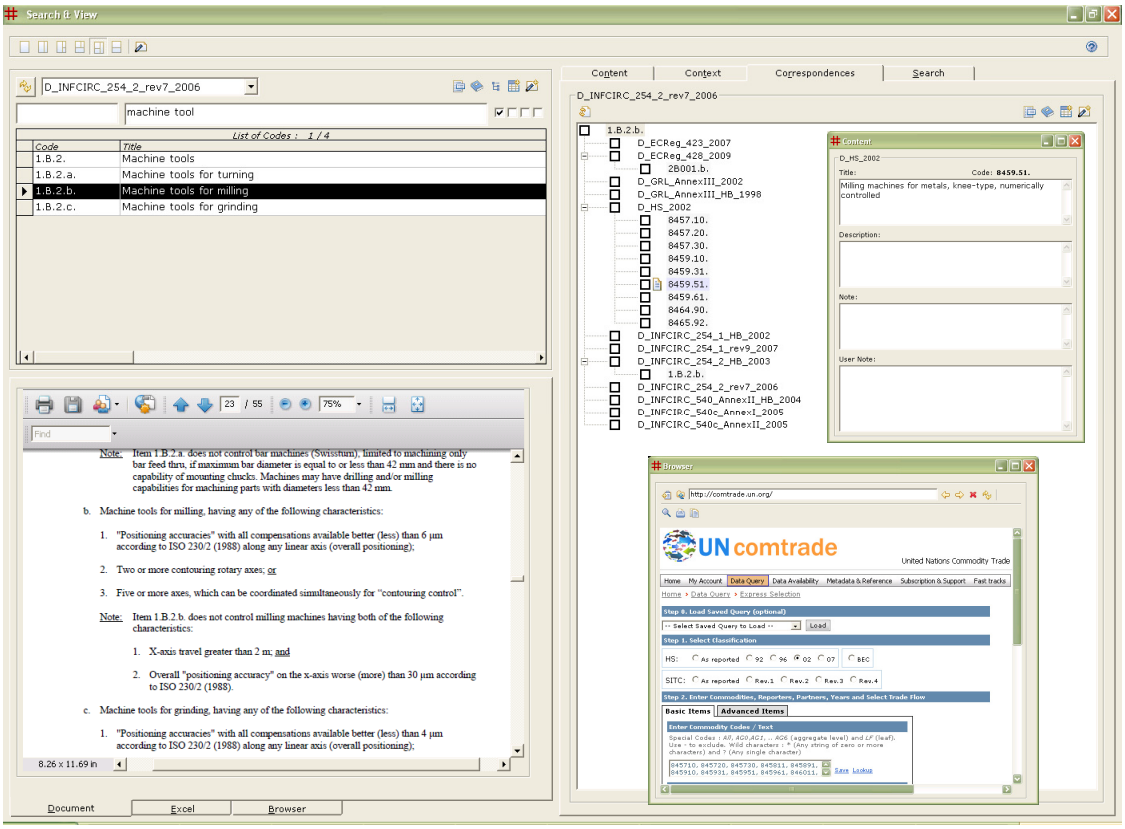

Figure 5 – TBT's *Search & View* interface.

7. TBT & related tools

TBT was designed and developed specifically to support work on nuclear trade analysis. To this end, the combination of tools offered by TBT is unique to the best of our knowledge. Some elements in TBT are related to other tools developed in the context of export controls. These are briefly reviewed hereafter. The majority of these are designed to inform EU exporters on legal restrictions that apply to trade. As such they support searching reference documents only in a few, selected directions. One tool (eCIT – SCORE) has been developed to support licensing and enforcement of export controls.

- **TARIC** [19] – Maintained by the European Commission (DG TAXUD) TARIC incorporates all EU legislation on trade, including EC Reg 428/2009 and EC Reg 423/2007. TARIC is a search tool published on a public web site. It can be accessed via TARIC codes, a subdivision of the HS. Given a TARIC code, the system retrieves the legislation that applies to the corresponding TARIC goods. This is the only search direction supported. TARIC does not contain technical information on items to which EU legislation applies.

- **Umschlüsselungsverzeichnis** [20] – The German export control authority BAFA publishes on its public web site a correspondence table between TARIC codes and items covered in EC Reg 428/2009. It is a PDF document organized by sections of the HS and intended to inform German exporters about restrictions that apply to the trade of items listed in Annex I of EC Reg 428/2009. The document does not contain technical information on the items covered and can be searched only by text. While the privileged order of consultation of the table is in the direction: TARIC code → EC dual-use regulation items, a full text search can be run on the set of PDF documents to derive the converse: dual-use items → TARIC for items of interest.

- **UK Goods Checker** [21] – The UK export control authority BERR publishes a web site to inform UK exporters on trade restrictions that apply to items listed in EC Reg 428/2009 and in national control lists. Access to the site requires registration. Legislation can be searched by text only. No reference is made to TARIC or HS codes. Some image data on controlled items is provided.

- **eCIT – SCORE** [22] – The U.S. Department of Energy (DOE) provides searchable online access to export controls lists as well as technical information on controlled items. Access to the site is restricted. This tool is meant for licensing and enforcement officials. Reference to the HS is made, but legislation or technical information on items covered in the system cannot be retrieved by HS codes. eCIT-SCORE appears to be the tool most related to TBT in that it connects various export control lists together and to technical information on items.

8. TBT's developments and perspective uses of global trade data

TBT is an application designed to support nuclear trade analysis for IAEA safeguards. TBT embeds a collection of reference documents listing items subject to controls and correspondence tables between related items, including Harmonized System descriptors. Trade analysis is then based on web global trade data retrieved by Harmonized System codes from data services outside TBT. TBT is already adopted by the IAEA Safeguards Information Management division to support the collection and analysis of trade data directly or indirectly relevant to safeguards. Planned developments and other perspective uses are discussed hereafter.

From the **software development** point of view, the main priority is to create TBT's *administrator* tool. The purpose of this tool is to manage and maintain over time the document collection and the correspondence tables between these documents. A desirable further development is to create a *web-based version of TBT*. This would enable a large community of users to work with a shared documents' database (instead of locally installed databases), providing users authentication, and controlling levels of accessibility to TBT's database content.

Concerning the **document collection**, it is foreseen to complete in the short term the itemization of the whole EC Reg 428/2009 Annex I on items from internationally agreed dual-use controls. These include the Wassenaar Arrangement, the Missile Technology Control Regime (MTCR), the Nuclear Suppliers' Group (NSG), the Australia Group (AG) and the Chemical Weapons Convention (CWC). Presently TBT embeds only items listed in the NSG Guidelines Part 1 [12] and Part 2 [6]. With items from all control regimes covered, TBT could support tasks such as the rating of items for *export licensing for all regimes*; especially if connecting Annex I items of the EC Regulation on dual-use to technical handbooks available from the control regimes to which EU Member States adhere. In this respect, it is foreseen to include in TBT at least the technical handbooks by the AG and the CWC. Likewise, it is planned to extend the document collection to lists (besides [17][18]) that restrict the export of specific items to particular destinations. These will include the European Regulation implementing restrictive measures against the DPRK [23][24].

Perspective uses of global trade data of relevance to JRC work pertain to the area of export controls.
In all countries the licensing of export-controlled items is the responsibility of national authorities. It is based on detailed information provided by exporters in license applications. It makes use of the technical expertise of licensing officers and a history of national trade on export-controlled items. However, there is a global perspective to export controls that goes beyond data and knowledge available at the single country level. This perspective could be provided by global trade data.
On the *enforcement* side of export controls, national customs authorities may need to consider global trade data to detect cases of *deflection* of trade. These are cases where nationally controlled items licensed for export to a given destination country are re-exported to a third country, thereby possibly circumventing national trade restrictions on the end-destination country. A second use in enforcement is to verify the effects of the implementation of sanctions related to embargoes.
At *policy* level, the effective implementation of export controls must facilitate low-risk trade while impeding trade connected to significant proliferation risks. In the EU one instrument to facilitate low-risk trade is the Community General Export Authorization (CGEA). CGEAs are configured as lists of controlled items and destinations for which export licences are not required. The decision about how to configure CGEAs is taken as EU level. Global trade data can help shape CGEAs on areas where facilitation of trade may be needed due to existing high-volume trade flows towards low-risk destinations.

In all cases sketched above, information derived from global trade data can play a role as presently no official data about the world trade on export-controlled items exists in formats that countries are ready to share. Yet global trade data can only give indications on trade covered by export control regulations due to approximations introduced by the use of Harmonized System descriptors.

Acknowledgement

The work here presented has been carried out within the European Commission (EC) Support Programme to the IAEA by the EC Joint Research Centre, Action on Information Analysis for Nuclear Security funded within the 7th Euratom Framework Programme. The authors wish to thank R. Chatelus (IAEA) and Cathy Pasterczyk (IAEA) with whom part of this work has been performed.

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Some ESARDA Parties' experience with Additional Protocol export control declarations

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Abstract:

The Additional Protocol to the Comprehensive Safeguards Agreement (INFCIRC/540) foresees a series of provisions which are the basis for Integrated Safeguards. For this reason, it is the legal bridge between traditional and strengthened safeguards, where export control officially appears as an additional barrier to proliferation.

Besides the introduction of short notice or unannounced random inspections, and complementary accesses, the AP also requires States to provide the IAEA with declarations concerning the presence of any of the fifteen nuclear fuel cycle activities listed in Annex I, as well as a summary of exports of items listed in Annex II, derived from the Nuclear Suppliers Group trigger list. Information about imports is instead due only upon specific request by the IAEA.

The paper will present the experience of some ESARDA members in complying with AP requirements for export control, outlining the possible issue of technical, information and timeliness nature, providing some suggestions for improvements. The paper will also address the significance of export control to the Information Driven Safeguards.

Keywords: Additional Protocol; Integrated Safeguards; Information Driven Safeguards; Export control; State's experience;

1. Introduction

Following the entry into force of the Non Proliferation Treaty (NPT) in 1970 [1], many steps were undertaken for the development of international nuclear safeguards and Comprehensive Safeguards Agreements (CSA), with the objectives of the prevention and detection of undeclared nuclear activities as well as of the diversion and misuse of nuclear materials from declared ones. Following that, the safeguards agreement between EURATOM and IAEA was established [2].

NPT Article III.2 requires safeguards as a principal condition of nuclear supply, with the following text:

> *Each State Party to the Treaty undertakes not to provide: (a) source or special fissionable material, or (b) equipment or material especially designed or prepared for the processing, use or production of special fissionable material, to any non-nuclear-weapon State for peaceful purposes, unless the source or special fissionable material shall be subject to the safeguards required by this Article.* [iii]

The need to interpret the meaning of the terms "*especially designed or prepared for*" components led to the formation of the NPT Exporters' (or Zangger) Committee. The resulting "Trigger List" (i.e. a list of equipment and facilities "triggering" the need for safeguards) and guidelines for the supply were communicated to Member States by the IAEA in INFCIRC/209 whose latest revision is reported in [3].

Following the Indian nuclear test, in 1974, the nuclear supplier states decided to form the "Nuclear Suppliers Group (NSG)" which also issued additional Guidelines in 1978, published in INFCIRC/254/Part 1 including an extended Trigger List [4].

Some years later in 1992, after the discovery of the covert Iraqi nuclear programme and realising the increasing role of dual use equipment, the NSG created an additional (Part II) set of guidelines for transfers of nuclear-related dual-use equipment, material and technology which could make a significant contribution to a nuclear proliferation programme [5].

For the same reasons, between the two key NSG dates, other international export control regimes were set up: the *Australia Group (AG)* in 1985, the *Missile Technology and Control Regime (MTCR)* in 1987, and the *Wassenaar Arrangement* (WA) in 1996.

2. The need for an Additional Protocol

The events in the 80's and 90's, culminating with the Iraqi programme and South African proliferation led also to the conclusion that the CSA were not giving enough mandate to inspections for the discovery of undeclared activities.

The work of the so-called 1993+2 programme resulted in the promulgation of the "Model Additional Protocol" (INFCIRC/540) in 1997 [6], supplementing the existing safeguards agreements by providing the IAEA with additional authority for both information and physical access to draw conclusions about the non-diversion of material declared through traditional safeguards and to verify the absence of undeclared nuclear material or activities.

The extended scope of safeguards under an AP gives broader IAEA access to sites and information related to nuclear fuel cycle research and development, paving the way to the so-called "integrated safeguards" system, based on classical accounting, a set of various types of inspection (short-notice, unannounced, complementary accesses) and the drawing of "broad conclusions" on State's absence of proliferation activities, based also on a series of non-classical indicators like export, import, satellite imagery, environmental analysis.

3. Export control and the Additional Protocol

Export and import of nuclear technology were (and increasingly are) seen as crucial indicators of proliferation activities. For this reason, AP's Article 2.a. requires that States:

> *..... shall provide the Agency with a declaration containing:*
> *(i) A general description of and information specifying the location of nuclear fuel cycle-related research and development activities not involving nuclear material…*
>
> and
> …
> *(iv) A description of the scale of operations for each location engaged in the activities specified in Annex I to this Protocol.*

AP annexes (I and II) were defined shortly after the detection of major clandestine activities (in this case in Iraq) and were relying largely upon previous work done e.g. by the Zangger Comittee and subsequently by the NSG. They were partially inspired by the "Voluntary Reporting Scheme" to IAEA introduced shortly earlier [7].

Annex I lists the following fifteen nuclear fuel cycle related activities:

i. The manufacture of *centrifuge rotor tubes* or the assembly of *gas centrifuges*.
ii. The manufacture of *diffusion barriers*.
iii. The manufacture or assembly of *laser-based systems*.
iv. The manufacture or assembly of *electromagnetic isotope separators*.

v. The manufacture or assembly of *columns* or *extraction equipment*.
vi. The manufacture of *aerodynamic separation nozzles* or *vortex tubes*.
vii. The manufacture or assembly of *uranium plasma generation systems*.
viii. The manufacture of *zirconium tubes*.
ix. The manufacture or upgrading of *heavy water or deuterium*.
x. The manufacture of *nuclear grade graphite*.
xi. The manufacture of *flasks for irradiated fuel*.
xii. The manufacture of *reactor control rods*.
xiii. The manufacture of *criticality safe tanks and vessels*.
xiv. The manufacture of *irradiated fuel element chopping machines*.
xv. The construction of *hot cells*.

Annex II lists instead the equipment instrumental to Annex I activities which can be derived from the Trigger List INFCIRC 254/Part 1, though in its 1995's version.

For Annex II's equipment and components, Art. 2.a.(ix) of the AP requires that the State:

> *…shall provide the Agency with a declaration containing the following information regarding specified equipment and non-nuclear material listed in Annex II:*
>
> > *For each export: the identity, quantity, location of intended use in the receiving State and date … of export;*
> >
> > *Upon specific request, confirmation as importing State of information provided by another State concerning the export of such equipment and material*

This means that the SSACs, or equivalent organizations depending on the countries' attribution of competences, are responsible also for retrieving this type of information and provide it to the IAEA along with the "classical" and other required declarations.

4. Export Control in the EU

As defined in [8], integrated safeguards entered in force in all EU-27 in 2010. All member states are hence due to provide their declarations to IAEA, via EURATOM or directly, depending on their agreements, including export declarations related to the AP.

At the same time, EU Member States must comply with the requirements set in EC Regulation 428/2009 of the Council of the European Union regulates the implementation of the export control of dual use goods (EC DU), their transfer, brokering service and transit in the EU [9]. This is a directly applicable regulation, which must be included into national legislation, implemented and enforced as set by the Community Customs Code [10] and its security amendments of 2005.

EC DU's Annex I includes the list of controlled dual use goods derived from all the international export control regimes (NSG, WA, AG, MTCR), as well as the Chemical Weapons Convention (CWC). Annex II is reserved to Community general Export Authorisations (for time being only one, but more are under discussion) and Annex IV includes a list of equipment for which an authorisation is due also for intra-EU export (transfer).

Hereinafter follows a summary of experiences of some EU member states participating in ESARDA, describing their organization and process for complying with export control requirements within the AP, outlining possible issues and improvements that will be discussed.

4.1 Spain

Export authorizations in Spain are given by the State Secretariat for Foreign Trade, whereas the responsibility for the Additional Protocol is with the State Secretariat for Energy. Both departments are in the Ministry of Industry, Tourism and Trade. The coordination is excellent, and every nuclear related export authorization is informed to the latter department. Moreover, the licences related to the defence and dual use material are discussed within an interdepartmental working group, formed by

representatives from different Ministries and Departments (Defence, Intelligence, Internal and External Affairs), where the Deputy Direction General for Nuclear Energy is present to provide his feedback.

The Additional Protocol entered into force in Spain, as a member of the European Union, on April 30th 2004. Within 200 days the first declaration concerning the period from 01/05/2004 to 31/06/2004 had to be submitted to the European Commission, including export declarations.

The first step that was taken was to inform all the Spanish nuclear industries and the related companies about the new commitment that has been established. In relation with exports, the different enterprises were asked to analyse the Annex II in detail to detect if they were obliged to declare any exported item. Once received the feedback, a number of enterprises and exports was identified to be included in the subsequent declarations.

A difficulty identified is the necessity to remind every quarter to the potential enterprises the obligation they have to send their information related with exports. In the last year this has gone more smoothly and it seems that it has, after six years, become a routine into the procedures of these enterprises.

Sometimes the bounds of the Annex II are not clear. There are some items that are not easy to rate under Annex II and it is not clear enough if they are subject to export control or not. In these cases a reasonable decision should be taken and the department asks the enterprise to analyse in detail the characteristics of their items in relation with the Annex II and to confirm if they are subject to export license and therefore to declaration.

Another difficulty is due to fact that export licenses are issued with a validity period before the export is actually executed, if at all. To complete this information, the department relies on the information sent by the enterprises, which have therefore to be fully aware and compliant. To make an accurate cross-check and complete export declarations, an official communication mechanism should be established to share the precise information owned by Customs department.

The importance of export control is significant if it is used to cross-check between the different States declarations, to match the export-import information sent by the different countries. However, the Ministry does not receive direct information about imports. In case the IAEA asked about a specific import, the information should be asked to the Customs department, and might take some time.

4.2 Hungary

The AP was signed by Hungary in 1998 and entered into force in April 2000. At that time, out of the States with considerable nuclear industry, only Japan, Australia and Canada had Additional Protocol in force. The initial Additional Protocol declaration of Hungary was sent to the IAEA in 2000. Since October of 2000 Hungary has submitted 11 comprehensive and 42 export declarations for the IAEA by the Hungarian Atomic Energy Authority (HAEA) acting as the safeguards regulatory body. After Hungary became member of the European Union in 2004, the multilateral safeguards agreement between the IAEA, the Euratom and the Member States came into force in Hungary on the 1st of July in 2007. Accordingly the State's obligation to fulfil the Additional Protocol provisions fell into shared competence. Being a non-side letter country, Hungary sends export declarations directly to the IAEA and in copy to Euratom.

Hungary engaged in the Zanger Committee's activities among the firsts in 1974, joined the Nuclear Suppliers Group (NSG) in 1985 and accomplished the presidential tasks of NSG in the period 2009-2010. National import/export control regime was established by domestic legislation in 1986 to harmonize with NSG Guidelines and control list, whereby the export of trigger list items became subject to authorization. After 1992, the control regime was changed to include the license requirements for the import of trigger list items and the export of dual-used items as well. By the time the AP came into force in 2000, national legislation had been changed to include notification requirements within 5 working days after the accomplishment of an export/import transaction. The HAEA informed enterprises with export capacities and import activities about the information provision necessary for AP declarations and the corresponding change in the national legislation. By this way HAEA could ensure that all information necessary for AP declaration was at its disposal as well as import information had it been asked by the IAEA.

EC DU Regulation [9] is currently implemented in Hungary by the Government Decree No. 13/2011(II. 22.) Korm. on the licensing of foreign trade with dual-use items and technologies, and the Hungarian Trade Licensing Office (HTLO) is the licensing authority for foreign trade matters. The specific requirements of the export, import and transfer of the nuclear and nuclear dual use goods are given by the Governmental Decree 263/2004 (IX.23.) Korm. For a license approval of nuclear items by HTLO the consent of the HAEA is necessary. HAEA performs the technical review of the applications and ensures full compliance with NSG Guidelines and safeguards requirements.

The Hungarian licensing system requires license for export and import of AP Annex II items with information provision obligations as explained above. The IAEA can therefore officially be informed about the accomplishment of the exports based on the information on the date of export provided by the licensee. There are some Trigger List INFCIRC 254/Part 1 items however, that are not subject to license for transfer between Members of the EU (some nuclear materials for example) as outlined by the Council Regulation and implemented in our national import/export control system. Therefore, as part of our safeguards licensing scheme, for nuclear materials whose transfer is not subject to the import/export control regime, a transport licence approved by the HAEA prior shipment is necessary.

The HAEA issues approx. 10-20 authorisations for import applications annually, involving fresh fuel assemblies for the nuclear power plant, smaller amount of other nuclear materials, measurement devices, equipment for the nuclear power plant and related technology, as well as intellectual products. The number of export applications is of similar magnitude, the majority of them having the subject of refuelling machines for reactors, their components and related technology. During technical review of the items that were subject to licence applications so far, the HAEA has not experienced real difficulties in rating these items.

For any nuclear items, whose import is subject to Government assurance required by the supplying country, the necessary commitment will be issued by the HAEA. The HAEA therefore performs 2-3 inspections of imported items annually to verify the declared end use and to have control over these items for their entire life cycle. Additionally exporters are also visited occasionally to brief them about the latest developments in the control regime and to discuss about their potential new activities and products that might be subject to control.

Developing a good relationship and establishing outreach activities with the potential exporters are a key factor in the implementation of an efficient control regime. This also implies for research institutions and universities with potential technology knowledge that might also be subject to licensing as intangible technology.

4.3 Sweden

Sweden signed the Additional Protocol, as a non side letter State, on September 22, 1998 and it entered into force on April 30, 2004. Thus, there was time to prepare routines for reporting the activities related to Annex I and export of items identified in Annex II of the Additional Protocol.

Export out of EU and intra-Community transfers of nuclear material and nuclear equipment were at the time already regulated by the Council Regulation 1334/2000 on export control now replaced by [9]. In the national implementation of the regulation, the Swedish Radiation Safety Authority (SSM) is the competent state authority to handle and decide on export and transfer applications of goods contained in Category 0 of Annex I to the Regulation. Category 0 contains nuclear material and nuclear equipment. Special cases that can be of principle interest can be handed over to the Government for a decision.

SSM is also authorised to request and give official assurances on the peaceful uses according to the NSG Guidelines for nuclear transfer. This is done for export out of EU if Sweden or Euratom have no bilateral agreement with the receiving country, giving equivalent assurances.

Sweden has since 1993 voluntarily provided the IAEA with information on approved export application on nuclear equipment. This can easily be done since SSM has a register of all applications. However, Sweden does not inform the IAEA on approved export applications on nuclear material or approved applications for the export of nuclear technology or software necessary for the ″development″,

″production″ or ″use″ of goods. Technology and software directly associated with any goods are controlled to the same level as the export of the item itself.

The voluntary reporting provided valuable experience in preparation for the Additional Protocol. The declaration according to the Additional Protocol, Article 2.A.(ix)(a) is however slightly different. The Additional Protocol requests information on exports out of EU, not on approved applications. This means that a follow up mechanism has to be introduced in the national regulations. A requirement is now included in the SSM regulation SSMFS 2008:3 requesting an exporter to inform SSM within one week of exports made. SSM collect this information and submit it on a quarterly basis to IAEA. Similarly as for the voluntary information given, Sweden does not provide the IAEA with information on exports of nuclear material or exports of nuclear technology and software. Information on exports of nuclear material is made to the IAEA via the nuclear accountancy system through Euratom.

Sweden has continued to voluntarily inform IAEA on approved export applications. After Sweden joined EU this information has been limited to exports out of EU. It can be questioned if this reporting should continue also with the Additional Protocol in place and declare the actual exports made. Sweden welcomes a discussion with other countries on this issue.

SSM has so far issued only so-called individual export authorizations for the export and transfer of nuclear equipment. In this way, SSM has full information on the type of equipment, number of items to be exported and the end user. This information is useful to follow up exports. The export regulation 428/2009 also gives the possibility to issue a “global” export authorisation, for example an authorisation for the export of a specific item to a specific end-user but without a limitation on the number of items. A global authorisation requires that the exporter has more extensive internal routines to keep track of the exports and to report it to the authorities.

If an application for export is refused the authorisation, the denial’s information is sent to NSG. Sweden regards that this information also covers the requirement to inform the member states of EU. Sweden is presently investigating if this information on denied exports could also be shared with IAEA (outside the Additional Protocol).

There is no import control of nuclear equipment in Sweden. SSM has no register of the entities importing nuclear equipment. For nuclear material there is a requirement according to the nuclear activities act for a permit to import. This is mainly to ascertain that the receiver has the relevant permits to handle the material. However, to date there has been no official request from IAEA for confirmation in connection with import to Sweden.

4.4 Finland

The Additional Protocol entered into force at same time as in other European Union countries, on 30.4.2004. As the Additional Protocol was seen to be coming, all the necessary preparations to fulfil Finnish obligations had been made during 2003-2004 to submit very first declarations in timely manner.

One of the most important tasks in the preparation process was to establish information flow procedures among the parties. It was seen important not only to fulfil the requirements but also to ensure transparency between the IAEA, EC and national authority. It was seen essential that as the State is responsible for nuclear activities without nuclear materials, the State shall also be responsible to collect and submit information about these activities. In Finland, the Radiation and Nuclear Safety Authority (STUK) is responsible of collection, inspection, evaluation and submission of declarations which are under State’s or common (with the EU) responsibility.

The most difficult task was to define the nuclear fuel cycle related research and development work where nuclear material is not involved. In connection with R&D review, operations of possible locations engaged in the manufacturing activities specified in the Additional Protocol Annex I as well as export and import activities in Annex II were examined. The conclusion was that there were not so many activities which may be considered possibly to be declared in the Additional Protocol 2a(i), 2a(iv) and 2a(ix)(a) declarations in Finland.

According to the Nuclear Energy Act (11.12.1987/990 with amendments) the person or undertaking who is going to import, possess or transfer equipment and non-nuclear materials have to apply license from STUK and those who practise nuclear fuel cycle-related research and development activities determined in Article 18(a) of the Additional Protocol will have to annually make notification to STUK. Manufacturing equipment and non-nuclear materials doesn't require license, but it is set in Nuclear Energy Degree (12.2.1988/161 with amendments) that person or undertaking who is going to manufacture those materials has to make notification about this to STUK. It is self-evident that STUK has to verify the correctness of the information it has received. STUK also makes annual review of the activities that may have to be reported according to the Additional Protocol.

To export the equipment and non-nuclear material identified in the Additional Protocol Annex II, the person or undertaking is obliged to follow the Act on the Control of Exports of Dual-Use Goods and apply for an export license from the Ministry for Foreign Affairs (MFA). Taking into account the obligations raising from the Nuclear Energy Act, STUK shall be aware of equipment or non-nuclear material subject of export license application. Customarily, MFA asks statements also from STUK about the export license application. If the export license is granted by MFA, there is also license condition set that of each export the licensee must make notification containing all the required information for the Additional Protocol declarations in two weeks after the event. STUK and MFA organise annual meetings with the Ministry of Employment and the Economy (MEE) on which export control matters of dual use items as well as other current safeguards issues are discussed thoroughly.

The initial AP declaration was submitted on 8.7.2004, and it consisted of all the Additional Protocol declarations required, including the first export declarations from 30.4.2004 to 30.6.2004 (second quarter of 2004). Until end of March 2011, altogether 27 export declarations have been submitted to IAEA by STUK. Most of these declarations have been "nothing to declare", 7 declarations have contained one export line entry each.

Developing a good relationship with potential exporters, importers and manufacturers as well as those who might carry out research and development activities is essential. Also good co-operation between the authorities is seen important in Finland, not only between STUK and MFA/MEE, but also with Customs and national security authorities.

The best way to reach success is the open and regular communication between all parties. According to our experience, knowing the responsibilities and exchanging the information openly is the most important duty in implementing the IAEA integrated safeguards.

4.5 Lithuania

The Additional Protocol for Lithuania came into force in July, 2000. In May, 2004 Lithuania became a member of the European Union (EU). The Community regime for the control of exports of dual-use items and technology has been fully adopted. In 2004's plenary meeting of Nuclear Supplier Group (NSG), Lithuania was accepted as the NSG member too. Since January 2008 the trilateral Safeguards Agreement and its Additional Protocol (between the non-nuclear weapon States of EU, European Atomic Energy Community and IAEA) superseded the bilateral ones between the Government of Lithuania and the IAEA. The interlink between the two issues has been established and maintained from the very beginning of their implementation in Lithuania.

The Ministry of Economy is the competent authority in Lithuania for licensing of activities with the strategic goods, i.e. export, transit and brokering of dual-use and military items. The inter-institutional group is formed by the different governmental institutions to assist Ministry of Economy for the review of the corresponding applications. The State Nuclear Power Safety Inspectorate (VATESI) contributes to the nuclear aspects.

Among its other duties, VATESI is also the authority responsible for the national supervision of safeguards application in the country. The export applications submitted to VATESI for review provide the initial information necessary in case of declaring exports to the IAEA under the Additional Protocol. The requirements issued by VATESI on "Nuclear Material Accountancy and Control and Provision of Information about the Activities in the Nuclear Energy and other related to it Fields" provides that within two weeks an exporter has to notify VATESI of the exact date the actual export has taken place.

However, with the Ignalina NPP still in operation, the nuclear items were predominantly imported into Lithuania. The supplier countries following the NSG guidelines would require the Government assurance for the trigger items of the NSG list. The Lithuanian Government Assurance would be co-signed by the Minister of Economy and the Head of VATESI. Therefore, in case the IAEA had requested pursuant to the Additional Protocol the confirmation of an import into Lithuania, VATESI would have been in position to do that. Furthermore, the Government's "Order on Control of Strategic Goods" obligated VATESI to conduct at least annually an inspection in order to check the use of imported nuclear items, for which the Government Assurance had been issued.

Since the final shut down of the Ignalina NPP at the end 2009, the dismantling activities on the NPP site are gaining momentum. Thus, the former importer, the Ignalina NPP, has become a potential source of nuclear trigger and dual-use items as during the decommissioning process it will be sought to sell equipment as still could be used or as scrap metal. Therefore, VATESI requested that items prior to being sold, especially invoking an international auction process, would be checked against the control list of Council Regulation, ditto of NSG and the Additional Protocol, and the export control procedures would be assured.

5. Discussion and conclusions

The experiences presented show how AP's export control requirements are fulfilled as the consequence of EC requirements included into national law and their implementation by appropriate procedures. On the substance there appears to be no particular problems, however some difficulties were outlined and open discussions proposed.

It emerges that the following elements are fundamental to an effective export control system and AP compliance:

- Awareness and information of the exporters on their duties
- Communication and exchange among the involved actors, i.e. export control authority, customs and nuclear authority, also within EU-27

In this respect, new or updated mechanisms should be established to have access to all the information in a precise and accurate way. This requires time and different stages so all the people under a country that deal with the information learn the importance of this issue. The authorities should moreover also carry out regular audits.

Sweden raised the point about the voluntary reporting scheme, questioning if it should be continued also with the Additional Protocol in place and consequent declaration to IAEA.

Linked to this and considering that proliferation threats always present, one can also ask whether the AP's export/import requirements are enough to provide the IAEA with sufficient declared data on activities and related trade. The number of "controlled" activities in Annex I and the associated items in Annex II could be expanded to better address the issue.

Since the definition of AP Annexes, the NSG has already modified the Trigger List of controlled equipment. NSG list Part 1, is now more extensive and including more items than AP's Annex II, still based on the older Trigger List's version of 1995. Moreover, the identified proliferation programmes outlined how dual use items, included in NSG's Dual-Use List (but also in other regimes) are functionally related to the nuclear fuel cycle.

An example is Annex I's Activity iii "manufacture or assembly of *laser-based systems"*: NSG Dual use list includes various elements of lasers systems that could be added to better describe the activity. As suggested already in [11], these consideration clearly support the fact that Annex II should be refreshed and completed both with an updated Trigger List and including Dual Use items to provide a more complete and up-to-date set of indicators.

Connected to this there is also the fact that the NSG has just started an extensive review of both Trigger and dual-use lists due to be completed in 2013. This review will result in modified controls, either in terms of additional items, in parameters' modifications or also decontrol of items.

The EU legislation will adopt these changes and incorporate them in Annex I of EC Regulation 428/2009. Updating AP's Annex II should hence be the logical consequence, to remove present and future inconsistencies. Indeed already now Member States' authorities and exporters have to work with two different lists of equipment: EC DU's one covering equipment that needs an export authorisation and AP's Annex II with items whose exports need to be reported to IAEA. This is clearly a difficult and confusing situation.

Updating Annex II of the Additional Protocol to be in conformity with the NSG lists, requires a specific decision, and would inevitably lead to a large increase of data to be retrieved and communicated. Expanding Annexes may indeed raise concerns linked to the increase in workload, adding up to the uncertainty about the effective export data correctness and completeness. This uncertainty should of course be kept to a minimum by first improving the communication and exchange mechanisms, as observed above.

Additionally, as already proposed in [12], considerations could be made about adding also import data related to Annex II to AP declarations.

The authors wish to get also other ESARDA Parties' experience and feed-back on this subject, as well as discussing the various points outlined and proposed.

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33rd ESARDA ANNUAL MEETING **Paper Number: 101**

Responsible behaviour of the dual use industry

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Abstract:

Export controls are mainly made for industry to comply with. Academia and its intangible technology transfer capabilities must not be forgotten, but without the supplies of in particular high-tech companies WMD and missile projects of sensitive countries are simply not possible. Neither do they have the technology, nor all the domestic manufacturing capabilities and skills to produce what is needed.

In recognition of its own responsibility, industry should not only pay due respect to all prevailing regulations, but also go beyond the stipulations of the laws. These rules often lack effectiveness, owing to a variety of fact: governments aim to avoid unnecessary burden for industry, the export control lists put a few fences only around certain technologies; catch-all clauses will often fail since exporters will hardly ever have a “positive” knowledge of a restricted end use; sensitive countries seek to buy equipment the technical features of which stay below the parameters set by the control lists, and they use front companies and networks, suggesting vague end uses or incorrect end use destinations.

Industry can contribute much to successfully combating proliferation, by raising internal awareness, establishing a company specific Internal Control Program, applying a voluntary self-restraint in export matters, and seek to establish information sharing with government authorities. Precious information, needed by government analysts and also the IAEA Safeguards Dept. would be lost otherwise.

1. Introduction

Ladies and Gentlemen,

First of all we would like to thank ESARDA for this invitation and kind introduction. It is a privilege to speak to such a distinguished audience.

While we prepared the papers for today, the devastating events in Japan caught up with us and since it has become clear that they will have serious nuclear effects on Japan and therefore the world, the people and finally the economy. We can all see that the global awareness for all issues connected with nuclear power is rising; rightfully so.

Yet, we are grateful to be employees of Leybold today, and not some thirty years ago, when nuclear proliferation was widely spread and the brand Leybold was tainted by dubious activities of a few individuals.

Most companies have some of this 'Jurassic Business' in their history. But the lessons we have learned is what counts today.

We learned that there are two major aspects to nuclear power:

1. Nuclear weapons, and a pressing demand to strengthen nuclear disarmament

2. Civil use of nuclear energy, meeting the world's rising need for electricity entailing the pressing demand for stronger and comprehensive safeguard systems and global efforts to combat nuclear proliferation.

Both fields of nuclear use are dependent on deliveries from industrial partners in both areas; the opportunities for nuclear proliferation are vast.

Of course, proliferation is – to some extent – hindered by export control regulations. For decades, the media are reporting on illegal shipments of high-tech commodities with mostly nuclear related end uses. In the worst case, the final destination is a clandestine, undeclared plant for **H**ighly **E**nriched **U**ranium (HEU) fabrication. While state actors drive their ambitious nuclear projects towards completion, it is often private companies trying to procure what is required for the nuclear activities. Those, at the delivering end of the supply chain, are also private entities, either middlemen or the manufacturers of products. The IAEA already has access to information related to attempted, intercepted and seized export shipments, and it knows about successful illegal transactions once these become known. However, there are many more traces out there, visible traces left by those who tried to procure high technology items. In many cases, states with clandestine nuclear ambitions have not the industrial infrastructure to produce all required goods within their own borders. Therefore, they have to target and contact potential suppliers in other countries. In recent years, many supplier countries have intensified their awareness-raising programs and intensified their industry outreach. There are companies receiving inquiries which they may not be able or do not want to serve. Maybe, because they see no chance to obtain an export license or, they don't want such business for ethical reasons. Such data, available in companies and only occasionally shared by companies with government authorities in member states, should be made accessible on a much broader front, and also be shared with the IAEA. In a large scale approach, this would lead to an ad hoc improvement of the verification efforts and help establish a unique early warning system. The IAEA needs effective help from more member states, primarily with regard to information sharing, as well as emphatic support for the IAEA Procurement Outreach Program. To share procurement data will not cost industry or respective member state authorities' any money; forwarding an email is as simple as a mouse click, and companies will feel rewarded by knowing they did the right thing. Supporting the global fight against potential nuclear terrorism and proliferation is in itself rewarding. And beside the IAEA, and our security authorities, the biggest winner of this simple approach will be us: the population of our global village.
Industry can indeed do a lot to slow down or even hinder illegal nuclear trade.
So, who is Oerlikon Leybold Vacuum and what do we do?

2. Export controls: a good tool in an almost empty tool box

The core of "awareness" related export control regulations is "positive knowledge" of the exporter about a WMD (weapons of mass destruction) or missile end use.

Only very few countries go beyond this and constitute license requirements if the exporter has “reason to believe” or “grounds for suspecting” any such end use. Countries initiating a WMD program will strive to cover their intention. The country will do everything it can to make sure that the program is camouflaged, most of the operations are run by intelligence services, and nothing leaks out. You will hardly ever be approached by any of the customers’ representatives explaining to you that there is a military end use, nor will you ever be shown any drawings or other documents that would enable you to file a license application owing to the “positive knowledge” that you have.

The goal of this speech is not to criticize export controls – they have their full justification and are as good as everything else man made. There are quite a few countries that work hard to introduce good and high level export controls, and other countries which work equally hard to help these countries to develop and implement this tool. Yet, like with any other tool, there is a limit to its functions and capabilities. Export controls are not enough to counteract proliferation purposes. It takes more and other efficient tools, as well as a dedicated group of mechanics interacting as a team with one common goal: to fight illicit nuclear trade. Today, we are focussing on a potentially powerful new team member: Industry. Hopefully, the aforesaid makes clear that the IAEA does not have the smallest interest to become yet another player on the export controls school yard. An additional export control agency will not help anybody, and also stricter export controls will not lead to greater success. We would just build higher hurdles for legitimate trade without moving a little step closer towards our goal, i.e. the early detection of unlawful programs or at least their late identification.

2.1 What is required must be imported

Industry and trade can play an important part in the prevention of proliferation of WMD and missiles. Proliferation is dependent on importing large quantities of high-technology components and products – so they have to be exported by the producing or selling companies. Trade always leaves traces that can be identified by industry, and so do attempts of illicit procurement. We still see many inquiries coming through already known channels and often from the same still existing front companies.

For us, as supplier of vacuum components, systems and spare parts, the ‘shopping list’ still contains the same items. Despite all efforts, there is still large-scale “procurement” out there for unlawful purposes and this reveals a very simple fact: The countries engaging in illegal programs mostly do not have the resources and capabilities to produce the required technologies domestically. They depend on a limited number of exporting companies in order to obtain what is required. This leads to an interesting question: What would happen if all potential suppliers refused to deliver? Or if many of these suppliers would be willing to share their insights on dubious inquiries and reported these voluntarily to government authorities and via these – to the IAEA?

2.2 Industry is the front line of defence

As a matter of fact, industry is in the front line of defence when it comes to stopping procurement for nuclear enrichment, WMD, and missiles. These trade traces, however, will hardly be available for governmental authorities unless the company, for whatever reason, chooses to communicate and actively forwards the information to the authorities.
Procurement for illicit programs starts just as any other trade process within the “Supply Chain”. It starts with a requirement, an inquiry to a potential supplier, leading to a quotation. Negotiations might follow, resulting in a customer order or contract, an order confirmation, followed by a production process. Then shipping procedures ensue, through a forwarding company, and it will finally involve the financial system for payments. Many actors in industry, trade, transportation and finance are involved. But the most suitable partner for the identification of what might be a suspicious or illicit requirement is the manufacturer and exporter of the goods involved. He has the best overall survey with regards to the entire transaction, the suitability of the product for the claimed end use and the judgement on its plausibility. May be there are protocols of “red flags” that would make the company suspicious, and a good internal compliance system would help the company to identify if identical or similar inquiries or orders were rejected. Often, similar requests come either from the same company to multiple affiliates of a producer, or, from a different entity in another country, but with the same technical specification and for identical quantities. Here is just a recent case:

There was an inquiry from an e-mail account in the UK following an entry in a US search registry into our US affiliated company. The products are standard items, no export licenses are necessary at all. Without the information of our German authorities that these items were on the shopping list of a certain country, we would not have noticed anything extraordinary. Alerted, we searched our data base and found those independent inquiries from several countries to several of our offices. Consequently, we did not quote, but shared all cases with the authorities, helping them to see the larger picture and to add several names to their data base.

Please keep in mind that vacuum is an indispensable enabler of many state-of-the-art production processes, but also ancient technologies like the electromagnetic isotope separation or first generation ultracentrifuges require still the same vacuum components. The same applies to processes in connection with missiles, be it the handling of propellants, the coating of electro-optic devices, the manufacturing of testing of gyros or the high altitude simulation. Vacuum is a “must”.

The inquiries we receive – and do not serve – have huge coverage. It could be just a set of spare gaskets for a vacuum pump delivered back in the 1980ies. If we allow the spare parts to reach their final destination, they will enable the operator to replace the gaskets and the pump will continue to work in a process in which we do not want it to work. We often hear questions from our sales force, whether we can perhaps define a group of harmless parts which they can sell without concerns, and without being so cautious. Looking at the seemingly harmless spare part or the standard product and the not so harmless process involved you will understand why the answer is NO.

There are hundreds of such suspicious inquiries and our company rejected business with a total value of major double digits of millions - voluntarily.

Our international export control group is working as what we call a “centralized detection hub”. We operate subsidiaries in 17 countries. We train the global sales force, service technicians, order management group, product managers and everybody else who might have customer contact.
To be clear about it: to combat illicit procurement, it is not enough to concentrate on a handful of ‘wrong doing’ countries. In brief, all countries qualify for diversion of equipment and illegitimate procurement, and this is one of the reasons why we centralized the matter. If we discover a particularly tricky or dangerous procurement attempt, we send a warning to our subsidiaries.

2.3 “Lost information” – unavailable for analysis

In spring 1992, the Leybold AG, that time a German high-tech group with then several thousand employees and two major hubs in Hanau (near Frankfurt) and in Cologne, introduced a voluntary self-restraint in export matters. It became known as “the LEYBOLD Charter”.

It demanded that all employees should strictly adhere to our company business ethics and export control procedures. The Charter also had and still has a clause that we do not engage in a business if we have – even after receipt of an end user declaration – continued concerns regarding the end use of the goods we are asked to deliver. You may imagine that after a short while we received a bunch of inquiries that were not to be turned into business. These inquiries included requirements from embargoed destinations as well as nuclear and missile programs in several countries.

The next slide shows how much information is available in industry and how much is lost, when this information remains in our company’s archives or ends up in the trash bin. Lost for government authorities and the IAEA. Authorities do have a certain access to information that may be relevant for the prevention of proliferation, and they have access to data they can get out of export license applications. But all the rest is unavailable, unless actively communicated and voluntarily shared by industry.

2.4 Early warning or late identification

During our trainings for subsidiaries and in particular during Export Control Outreach seminars in a variety of countries, we found out that there is often little or no knowledge on laws and regulations which prevail in the respective countries. Many companies have very little awareness; do not screen their product portfolio against the current export control lists or against Sanctioned Party lists.
It has to be assumed that there are still many shipments that go without the necessary license, and that the exporter simply does not know that there is a license requirement. There are, of course, also cases in which an exporter chooses to ship without license deliberately, be it by means of false customs tariff numbers, product description, incorrect country of final destination or whatever he deems necessary to execute on this business. The largest group by far is the huge quantity of inquiries – normal and illegal ones - that exporting companies do not execute on, for very different reasons.

Yet, these companies are often able to discern between a normal inquiry and a potentially problematic one, and the latter are exactly the ones that government authorities should be informed about. If government authorities and the IAEA had better access to these many puzzle pieces, they could certainly analyze these and get a better feel for newly developing nuclear ambitions of countries that have not shown any such interest so far.

In any case, industry-government information and fearless communication with government officials is an indispensable verification tool that effective Safeguards need. Other tools are already in use, such as physical inspection, surveillance cameras, environmental tests and satellite imagery. Industrial information would just be another tool in the tool box.

2.5 Winners, losers and indispensable prerequisites

We do not try to persuade other companies to follow our example; industrial leaders have to come to their own conclusions regarding their ultimate social responsibility and to reduce the threat caused by the world's most dangerous weapons. What separates us from other species is the free will to decide to do "good," to act socially responsible. Is it acceptable to execute on business in the grey area, where the knowledge for unlawful purposes is perceived, but where laws do not work effectively enough? Is generating share- and stakeholder value limited to the fiscal bottom line? Our stakeholders rely on us to maintain certain, if not the highest, moral and business ethics – for ourselves, the social surrounding and for our future.

From our experience, the last thing that exporting companies will want to do, is to create problems for themselves. No one is really looking forward to a visit of officials because they have a legal obligation to investigate the company after they have learned about a potential violation.

If we want these companies to share their information, we have to create a culture of mutual trust and understanding. In particular, companies must be enabled to share such information without having to be afraid that the voluntarily supplied information will be turned and used against them. Some companies will already have achieved a high level of social responsibility, adhere to compliance guidelines and might easily be won over to participate. For other companies, enticement programs might be conceivable, for example with incentives from the financial communities, or with specialized export procedures.

Yet, we have to keep in mind that the work with industry requires a legal framework, manpower and the right skills including technical understanding. Authorities may face new challenges regarding the education of their officials, and may soon come to a point where they obtain much more information than they can digest. Moreover, the IAEA has a chronicle disease, the shortage of funds and human capacity. It should be ensured that there is enough manpower and other resources to manage the information flow in a timely manner, and there should be a broader stream of shared information from many more member states.

Nuclear security, global stability and world peace are not available for free – if this is our belief and if we think that – after the globalization of proliferation – also counter proliferation should indeed become global, we ought to have more tools in the tool box. We should then take the necessary next steps, to create appropriate working conditions and provide resources necessary.

Dear Ladies and Gentlemen, let us finish by quoting famous physicists:

Concern for man and his fate must always form the chief interest of all technical endeavors.
Never forget this in the midst of your diagrams and equations.

Albert Einstein

Thank you for your attention!

14 Environmental Sampling – Particles

Method development for analysis of single particles for safeguards purposes

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Abstract:

This paper describes a method developed for the particle analysis of safeguards swipe samples. The method consists of several, individual procedures. For the identification of single particles, solid state nuclear detectors were used. For the localization of them a calculation method based on coordinate transformation was adapted. The location of the particles was determined with an accuracy of better than 20 µm in case of particles with 80...120 µm size. The U isotopic composition of the located particles of interest was determined by laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS).

Keywords: particles; LA-ICP-MS; U isotopic composition

1. Introduction

Swipe sampling has been routinely used as an effective inspection tool by the International Atomic Energy Agency (IAEA) to verify the presence or the absence of undeclared nuclear materials and activities [1]. Recently, particle analysis has been in focus besides bulk analysis since the latter provides information only about the average composition of the samples however, the artificial fissile material in the environment mostly occurs in the form of solid, radioactive particles surrounded by a large number of particles containing fissile material of natural origin. The results (such as isotope ratios of actinides) given by the analysis of different single particles offers a great opportunity: various origin scenarios of particles can be distinguished in the same swipe sample. Moreover, the more precise information can improve the verification process of IAEA safeguards.

The Hungarian Atomic Energy Authority (HAEA) has also been using swipe sampling for national safeguards purposes. Swipe samples collected by the HAEA inspectors in the nuclear facilities of Hungary are analyzed as bulk materials at the Institute of Isotopes (IKI). This study focuses on the development of a method to determine the isotopic composition of U in single particles originating from swipe samples which can supplement the routinely used bulk analysis of swipe samples at IKI.

In the present paper, procedures developed and adapted for the identification of single particles using solid state nuclear track detectors, the localization and the analysis of the identified particles using LA-ICP-MS are to be discussed.

2. Experimental

2.1. Instrumentation

The mass spectrometric analysis was carried out using an Element 2 ICP-MS (Thermo Electron Corp., Germany) with magnetic sector field and single electron multiplier. The laser ablation was carried out using an UP-213 laser ablation system (New Wave Research, Freemont, USA) equipped with a Nd:YAG laser at a wavelength of 213 nm (pulse duration < 4 ns).

2.2. Samples

Test particles containing U were made of anion exchange resin (BioRad Ltd., USA) beads to simulate particles for testing the adapted and developed procedures. Uranyl chloride (Reanal Ltd., Budapest) was loaded onto anion exchange resins. Each anion exchange bead with an average diameter of 140 µm contained approximately 13 mBq ^{238}U (1 µg U).

Particles originating from confiscated nuclear fuel pellets with different U isotopic composition were also analyzed. Particles were removed from three pellets using adhesive, transparent tapes which were used afterwards as sample holders for the further examinations. The uranium in the samples had different enrichment: depleted (DU), natural (NU) and low enriched (LEU) uranium. Pellets from the same batch were analyzed during a joint analysis project involving the JRC Institute of Transuranium Elements (ITU) and the Institute of Isotopes using different analytical methods, such as multi-collector ICP-MS (MC-ICP-MS), thermal ionization mass spectrometry (TIMS), solution nebulization ICP-MS (SN-ICP-MS), high-resolution gamma spectrometry (HRGS) and LA-ICP-MS [2]. The results of this project were considered as reference values.

The size of the investigated particles varied between 80...120 µm and 9...247 µm in case of test and pellet particles, respectively.

2.3. The procedures of the method for particle analysis

2.3.1. Identification of particles containing alpha emitting material using solid state nuclear track detectors

The procedure for the identification of particles containing alpha emitting material is based on alpha track analysis (Figure 1) using CR-39 type solid state nuclear track detector (RadoSys Ltd., Hungary).

Nuclear track detectors were placed on the sample holders containing particles. The detectors were exposed to the samples for several days depending on the type of the sample. After unpacking the samples, the detectors were rinsed with ethyl alcohol, etched in 6.25 M NaOH at 90 ± 3 ºC for 4 h, rinsed with distilled water and dried in air. Alpha tracks were observed with optical microscope of New Wave Research UP-213 laser ablation system.

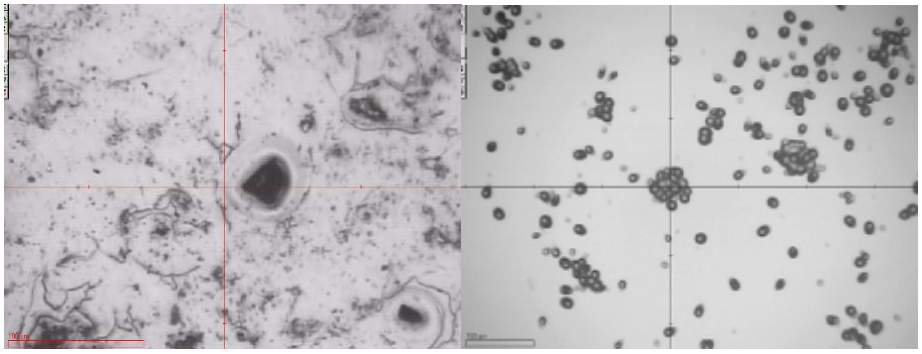

Figure 1: A particle originating from nuclear fuel pellet and the image of the alpha track formed by it (in the cross-hairs)

2.3.2. Localization and re-localization of the identified particles

The exact location of single particles containing alpha emitting materials is needed for further examination. The so-called 6-point algorithm calculation method was used for the determination of the particle coordinates in the coordinates system of the given microscope [3].

Three reference marks were placed on the sample holder (H_1, H_2, H_3) and three reference marks were defined on the nuclear track detectors (D_1, D_2, D_3), as well. According to the coordinates of the

reference marks and the coordinates of the nuclear tracks (T), the coordinates of the particles (P) can be calculated. (Figure 2)

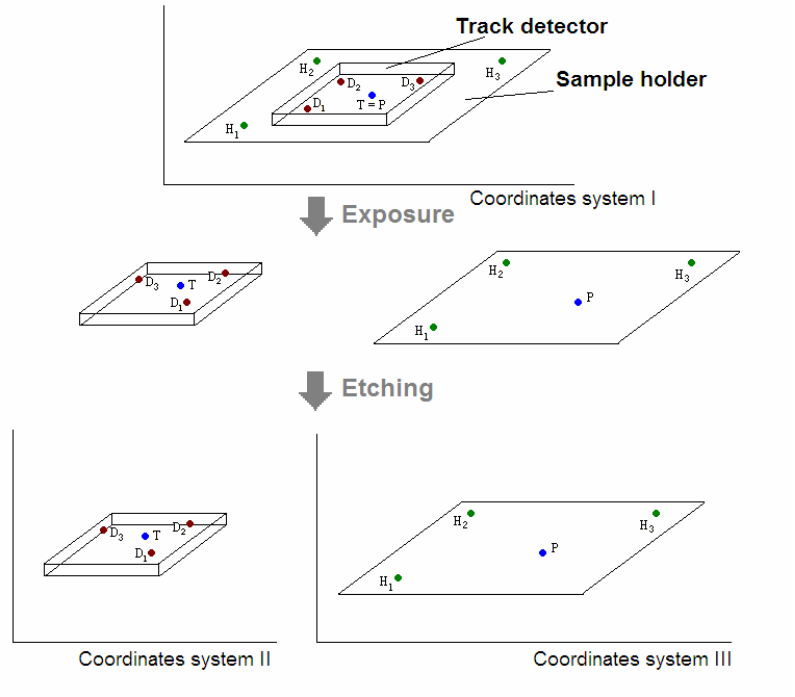

Figure 2: Determination of the coordinates of a particle using 6-point algorithm

2.3.3. Determination of the U isotopic composition of identified and localized particles by LA-ICP-MS

The identified and localized particles were analyzed by LA-ICP-MS. A NIST 612 glass reference material (NIST, USA) was used for the optimization of the LA-ICP-MS system. The achieved sensitivity was approximately 10^5 cps for 37.1 mg/kg ^{238}U. The mass bias factor was determined by the measurement of the ^{235}U/^{238}U ratio of a highly enriched U-oxide pellet (from an interlaboratory exercise of Nuclear Smuggling International Technical Working Group (ITWG), 2001). Low (R=300) and medium (R=4000) mass resolutions were applied. The used laser ablation technique was based on the method described by Z. Varga [4]. Spot ablation (spot size (laser beam diameter): 40 or 60 µm, laser energy flux: 0.04 mJ/cm^2, repetition time: 10 Hz) was used for the laser ablation.

The intensities of the U isotopes were recorded in the time resolved mode of the ICP-MS during the laser ablation of the particles. Three measurements were applied on each sample. The background level was measured in 25-35 measuring points by starting to record the chromatograms before starting the laser ablation. The U isotope ratios (^{235}U/^{238}U, ^{234}U/^{238}U, ^{236}U/^{238}U) were calculated from the average of a given time interval of the U isotope signals after the background correction. The U isotopic composition (the mass ratio of one isotope relative to the sum of all isotopes of the element) of the particles was evaluated, as well. The results for particles originating from the U oxide pellets were compared to the results of the joint analysis project mentioned below.

In case of the mass spectrometric analysis of the joint analysis project, a part of the pellets were dissolved and treated by radiochemical methods prior to the measurements. During the LA-ICP-MS measurement the whole pellet was placed in the chamber of the laser ablation system and line scan ablation was applied.

3. Results and discussion

3.1. Identification and localization of single particles

The parameters of alpha track analysis (exposure time, etching time, etching temperature) were optimized to achieve as precise localization as possible. In case of single particles the nuclear tracks form clusters. The exposure time is needed to allow for each cluster to have enough single tracks to be able to define precisely the center of the cluster. The optimal exposure time depends on the size of the particle and the alpha emitting material content. Five and 4 days were the optimal exposure times in case of the U test and the nuclear fuel particles, respectively. After the exposure the alpha tracks had to be enlarged since the alpha particle of U produces nanometer size tracks. The etching with the above mentioned etching parameters could enlarge the tracks to a diameter of approximately 30 µm.

The calculation methods for localization and re-localization of the identified particles were also tested. Test particles were fixed onto sample holders and alpha track analysis was executed. Six-point algorithm was used to calculate the coordinates of the particles, and then the center of the particle was determined. The difference between the calculated and the exact coordinates was defined as the precision of the calculation method. In case of re-location the same sample holders were examined in different positions or instruments. The achieved precision of the 6- and the 3-point algorithm was 17 µm and 13 µm, respectively in case of 80...120 µm particle size.

3.2. Determination of the U isotopic composition of particles by LA-ICP-MS

The described method was applied to U test and nuclear fuel pellet particles. Figure 3 shows the typical laser ablation signals of the different samples. The applied, low laser energy resulted in a relatively steady signal in logarithmic scale.

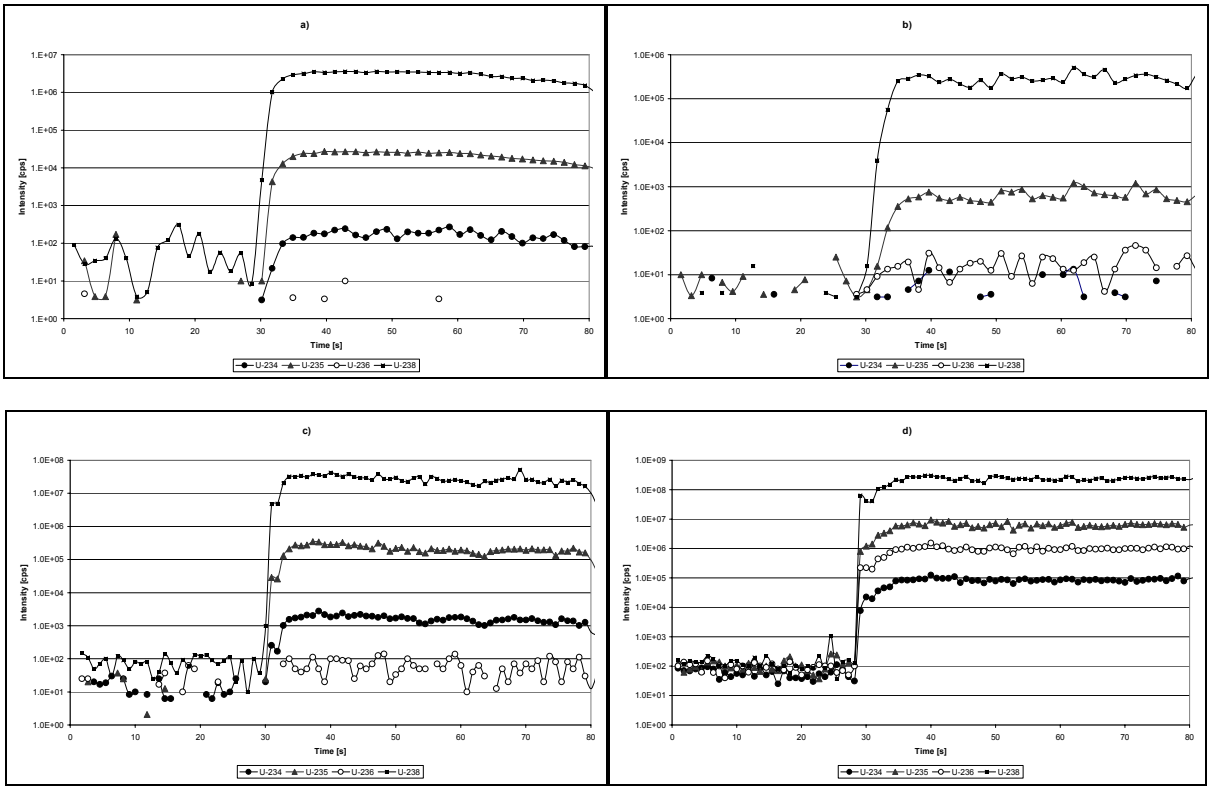

Figure 3: The chromatogram of a LA-ICP-MS measurement of single particles a) U test particles (d = 90 µm), b) DU particle (d = 9x18 µm), c) NU particle (d = 19x19 µm), d) LEU particle (d = 40x70 µm)

Natural isotopic U composition was predicted in case of U test particles since natural composition of the uranyl chloride was assumed. Typical results are shown in Table 1 and compared with the values

of the isotopic composition of natural U recommended by IUPAC (International Union of Pure and Applied Chemistry) [5]. The results agreed well with the values of IUPAC.

	IUPAC [%]	U test A [%±1σ]	U test B [%±1σ]	U test C [%±1σ]
^{234}U	0.0055±0.0005	0.0055±0.0004	0.0055±0.0005	0.0053±0.0002
^{235}U	0.7200±0.0012	0.7158±0.0166	0.7106±0.0354	0.7441±0.0482
^{238}U	99.2745±0.0060	99.2787±0.0168	99.2787±0.0358	99.2506±0.0482

Table 1: The calculated isotopic composition of the U test particles

The calculated U isotopic composition of the nuclear fuel pellet particles are shown in Table 2-4 and compared with the results originating from the joint analysis project mentioned above (marked us 'Ref'). The lateral dimensions of the analyzed particles are also listed in the table. In most cases good agreement can be observed between them even within 1 σ. According to the measured $^{235}U/^{238}U$ isotope ratios (Sample batch DU: 0.00277 ± 0.00015, Sample batch NU: 0.00706 ± 0.00032, Sample batch LEU: 0.0259 ± 0.0010), depleted, natural and low enriched U could be unambiguously distinguished.

	Ref DU [%±2σ]	DU-1 [%±1σ]	DU-2 [%±1σ]	DU-3 [%±1σ]
^{234}U	0.0014 ± 0.0002	0.0016 ± 0.0008	0.0020 ± 0.0004	0.0014 ± 0.0001
^{235}U	0.2635 ± 0.0018	0.2565 ± 0.0316	0.2934 ± 0.0290	0.2780 ± 0.0074
^{236}U	0.0058 ± 0.0072	0.0046 ± 0.0023	0.0067 ± 0.0015	0.0060 ± 0.0002
^{238}U	99.7307 ± 0.0019	99.7373 ± 0.0321 (9×18 µm)	99.6979 ± 0.0295 (31×54 µm)	99.7147 ± 0.0075 (150×170 µm)

Table 2: The calculated isotopic composition of particles originating from the nuclear fuel pellet containing depleted U

	Ref NU [%±2σ]	NU-1 [%±1σ]	NU-2 [%±1σ]	NU-3 [%±1σ]
^{234}U	0.0053 ± 0.0002	0.0052 ± 0.0006	0.0043 ± 0.0004	0.0052 ± 0.0001
^{235}U	0.7107 ± 0.0011	0.7054 ± 0.0232	0.7138 ± 0.0619	0.6846 ± 0.0700
^{238}U	99.2838 ± 0.0013	99.2892 ± 0.0235 (19×19 µm)	99.2817 ± 0.0627 (19×19 µm)	99.3097 ± 0.0709 (247×247 µm)

Table 3: The calculated isotopic composition of particles originating from the nuclear fuel pellet containing natural U

	Ref LEU [%±2σ]	LEU-1 [%±1σ]	LEU-2 [%±1σ]	LEU-3 [%±1σ]
^{234}U	0.0349 ± 0.0010	0.0492 ± 0.0278	0.0340 ± 0.0006	0.0329 ± 0.0012
^{235}U	2.5167 ± 0.0048	2.6041 ± 0.2485	2.4670 ± 0.1301	2.4662 ± 0.0889
^{236}U	0.4663 ± 0.2416	0.4170 ± 0.0540	0.4200 ± 0.0170	0.3640 ± 0.0070
^{238}U	96.9816 ± 0.0073	96.9295 ± 0.2575 (9×16 µm)	97.0787 ± 0.1322 (40×70 µm)	97.1370 ± 0.0900 (40×124 µm)

Table 4: The calculated isotopic composition of particles originating from the nuclear fuel pellet containing low enriched U

4. Conclusions

The method developed for the analysis of single particles in safeguards swipe samples for national safeguards purposes consists of several, individual procedures. Particles containing alpha emitting materials can be identified using solid state nuclear detectors. The location of the identified particles can be determined with an accuracy of better than 20 µm in case of particles with 80...120 µm size. The U isotope ratios and isotopic composition of the particles of interest can be determined by LA-ICP-MS with high accuracy.
The procedure has to be further developed for the effective removal of particles from swipe samples. Further improvements are also needed to be able to analyze particles under 10 µm size by LA-ICP-MS.

5. Acknowledgements

This study was financially supported by the Hungarian Atomic Energy Authority. Prof. Zoltán Szatmáry (Institute of Nuclear Techniques, Budapest University of Technology and Economics) and Dr. Árpád Vincze (Hungarian Atomic Energy Authority) are thanked for helpful discussions. The authors are grateful to Erik Hulber (RadoSys Ltd) for the nuclear track detectors.

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Improved uranium particle analysis using large-geometry SIMS

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***Abstract*:**

Large Geometry Secondary Ion Mass Spectrometry (LG-SIMS) is a new tool being applied by the International Atomic Energy Agency (IAEA) to the analysis of uranium particles from environmental samples for safeguards purposes. LG-SIMS provides a ten-fold increase in sensitivity, simultaneous collection of the uranium isotopes, and separation of interferences, leading to higher accuracy data that contribute to strengthening safeguards conclusions about the nuclear activities taking place in a State under Safeguards. Experience gained through analysis of 16 samples has led to a number of observations. Better quality of SIMS screening data enhances the selection of particles for accurate microprobe analysis in order to fully characterize the nuclear activities at a facility. Further, the efficiency of the system allows for more particles to be analyzed per sample with the same effort. These advantages allow reliable determination of the most depleted particles in a sample. Improvements in data for U-234 and U-236 permit deconvolution of different nuclear components with discrimination equivalent to that provided by the high accuracy technique available from the Network of Analytical Laboratories (NWAL), FT-TIMS (Fission Track-Thermal Ionization Mass Spectrometry). High precision of U-235/238 measurement reveals that variability in isotopic composition within a single particle is common for a number of samples. Performing uncertainty estimates according to the Guide to the Uncertainty of Measurements is critical in driving further improvements in measurement technique for LG-SIMS and thus efficiently deriving the maximum information from each sampling effort.

Keywords: analysis; uranium; particles

1. Introduction

The International Atomic Energy Agency (IAEA) collects environmental samples to verify the completeness of a State's declaration of their nuclear activities. One type of analysis involves measuring the isotopic composition of individual actinide particles, typically collected on a10 x 10 cm cotton swipe. As a part of the Network of Analytical Laboratories (NWAL) of the IAEA, the IAEA's Environmental Sample Laboratory (ESL) uses vacuum extraction of the particles onto a glassy carbon planchet followed by Secondary Ion Mass Spectrometry (SIMS) to determine the uranium isotopic composition of individual particles. The abundance of particles at different enrichment grades in U-235 is a characteristic measure of the activities at a given facility, while the concentrations of the minor isotopes (U-234 and U-236) provides additional information. Particle analysis, whether by SIMS or by Fission Track- Thermal ionization mass spectrometry (FT-TIMS) in the NWAL has proven to be a powerful tool contributing to the Agency's ability to draw strong safeguards conclusions.

The increasing complexity of safeguards goals over the last few decades has created the need for new methods of gathering more complete information about nuclear activities States. The use of the abundance of U-234 (verification of sources of uranium), and U-236 (evidence of exposure in a reactor) has the potential to provide additional information, but the low abundance of these isotopes makes the measurements technically challenging with conventional SIMS techniques.

M. Hedberg and colleagues [1, 2] have shown proof of principle that the recently developed “large geometry” (LG-) SIMS provides significant improvements in measurements of U-234 and U-236. The Agency completed acceptance testing of an LG-SIMS (model CAMECA IMS 1280) in July of 2010. In this paper we report on our first experience in using this instrument for safeguards samples and the benefits that it is bringing to the measurement of these samples.

2. LG-SIMS Characteristics

The much larger LG-SIMS (compared to the Agency’s Cameca IMS 4f SIMS) provides three major advantages. First, the instrument operates at high resolution, thereby separating out interfering species that mimic U-234 and U-236 (e.g., Pb-Si and Pb-Al). Eliminating these interferences solves past problems with false positives, generating much more reliable data. Second, the larger size allows enough space for simultaneous high sensitivity detectors for masses 234, 235, 236, 238, and 239 (U238-H or Pu). Simultaneous collection of all ion signals eliminates the problem of interpolation of varying or unstable signals as the particle is consumed during the microprobe measurement on a single detector [3]. This feature also ensures collection of more of the uranium signal, and combined with the higher overall sensitivity of the instrument leads to the third advantage, which is that the total signal collected from the same size particles is approximately ten times higher than with the Agency’s 4f SIMS. All of these advantages lead to more accurate isotopic measurements, especially for the U-234 and U-236.

Figure 1. The IAEA’s Large Geometry SIMS, being operated by an Agency staff member at the CAMECA factory.

3. Particle location

An advantage of the SIMS technique is that it can be used to rapidly locate a low small number of micron-sized uranium particles admixed with the dirt in an environmental swipe sample. Higher sensitivity for all isotopes enhances the quality of screening data collected with LG-SIMS. At ESL, the vacuum impactor

technique is used to transfer a portion of the sample from a cotton swipe onto a 2.5 cm diameter glassy carbon planchet. This planchet is rapidly and non-destructively scanned in the LG-SIMS over a series of 250 micron square fields for the presence of uranium isotopes. Specialized software ("Automated Particle Measurement" or APM programme from CAMECA) locates and estimates the uranium isotopic composition for particles within each field. Particles are then selected from this "screening" list for destructive microprobe analysis to accurately determine the uranium isotopic composition of a representative subset of the particles in a sample. Figure 2 shows a comparison of screening data with microprobe analysis results for U-235 abundance for the LG-SIMS. The data are reasonably well correlated. Figure 3 shows that past problems with isobaric interferences for data gathered with the 4f SIMS have been eliminated, considerably increasing the utility of this data set for tracking the pattern of nuclear activities associated with a facility at a given point in time.

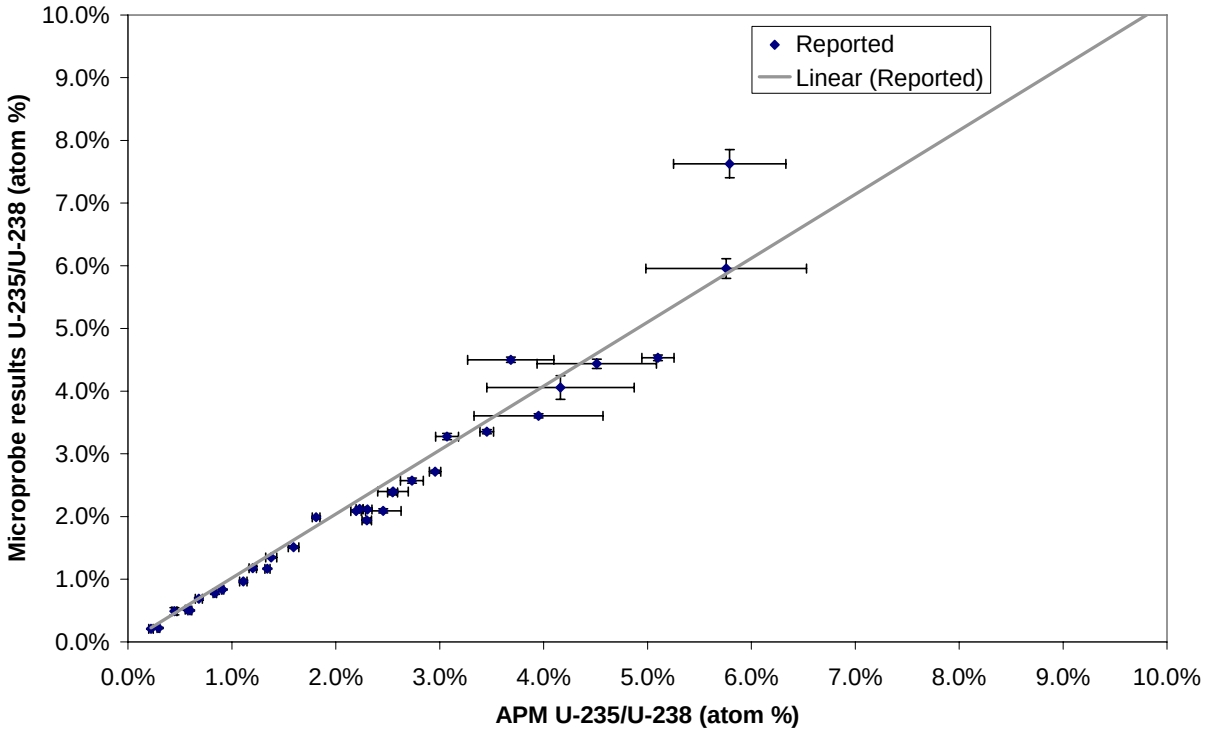

Figure 2. Screening data (generated using the APM software) vs microprobe data from LG-SIMS. Uncertainties for APM screening data reflect counting statistics only.

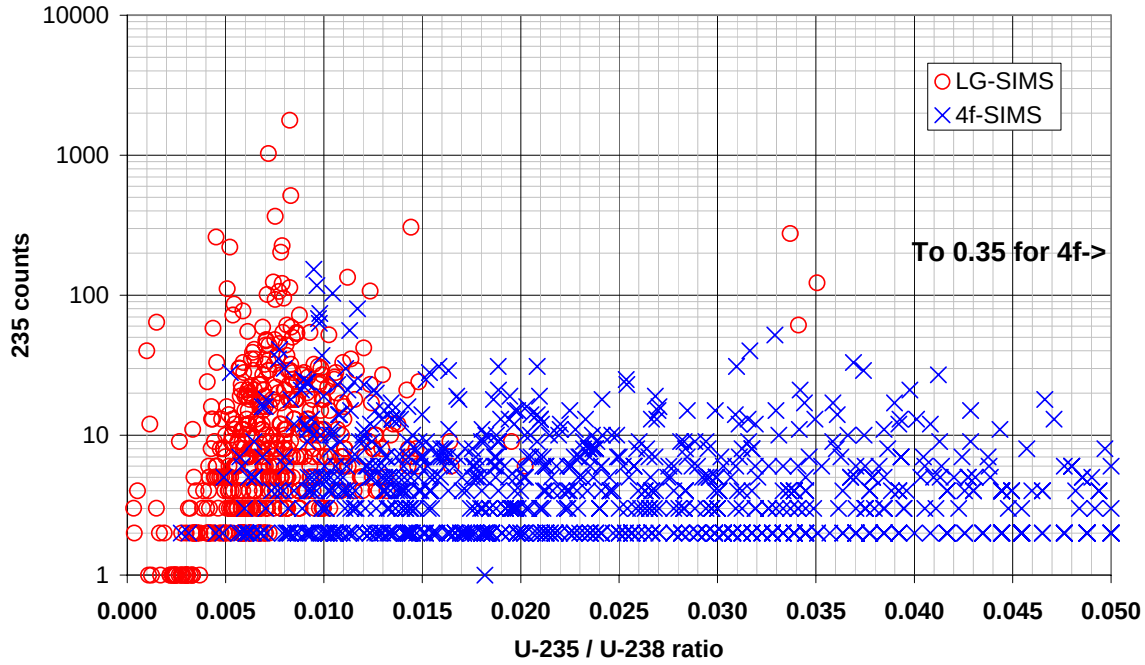

Figure 3. Screening data for the same sample planchet collected on both the 4f SIMS and the LG-SIMS. Many 4f data values are biased high due to isobaric interferences.

Obtaining high quality screening data is a key to selecting particles that represent all of the nuclear processes at a facility, thereby helping to achieve the safeguards goal of verifying the completeness of declared nuclear activities. Due to higher sensitivity, the new LG-SIMS adds a new capability: use of U-234 and U-236 screening data to select particles for detailed microprobe analysis. Figure 4 shows screening data from a case with unusual variability in the uranium data, where the high quality of the screening data was key to full characterization by LG-SIMS of the different nuclear processes affecting this sample. Another advantage observed for the LG-SIMS in the case of a complex sample is that microprobe analysis for each particle is much faster (8 vs. 45 minutes for the 4f SIMS protocol). This improvement makes it practical to collect data for many particles from a sample (up to 80), as needed.

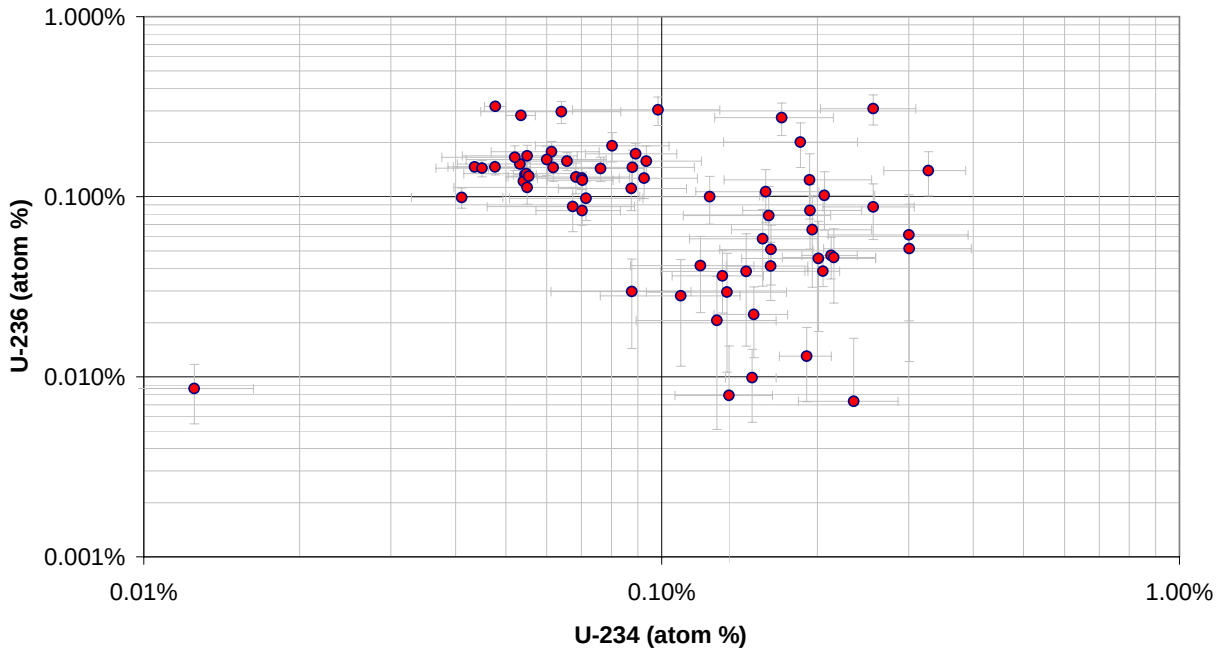

Figure 4. Screening data from LG-SIMS can also be used to search for different nuclear processes based on U-234 and U-236.

The high quality of the screening data may make it possible to use these data alone to monitor changes in activities at selected facilities, thereby increasing the efficiency of data collection. An updated version of the APM particle location program, based on customer feedback (e.g., [4] and IAEA experience), is in the testing phase and shows promise for improving performance at low 235 abundance (depleted Uranium) and discriminating between closely spaced particles. These improvements should lead to more accurate screening data with less post-processing work by the analyst.

4. Comparison to other techniques in the NWAL

4.1 Conventional SIMS (4f)

As mentioned above (Fig. 3), higher sensitivity and better discrimination against isobars lead to better data quality compared to previous SAL measurements using a 4f SIMS.
Figure 5 shows a comparison of data for U-234 and U-235 abundances from a sample analyzed by SAL on both the 4f and LG-SIMS. This sample represents the case of a low abundance of relatively small particles, which was relatively clean (low interferences). It was possible to locate many more particles with the LG-SIMS than the 4f SIMS, leading to an enhanced characterization of the sample. Better imaging and sensitivity may also reduce the effect of contamination by natural uranium background.

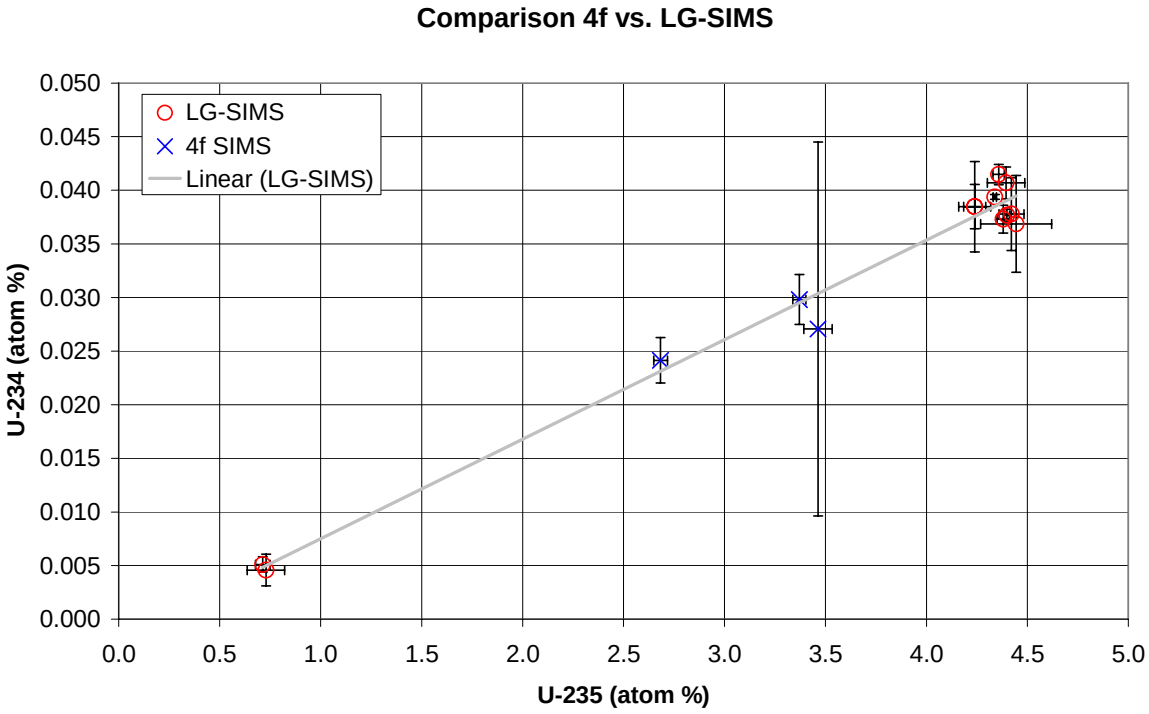

Figure 5. Comparison of LG-SIMS and 4f data for a sample with sparse, small particles. Data from LG-SIMS better defines sample and enrichment end members.

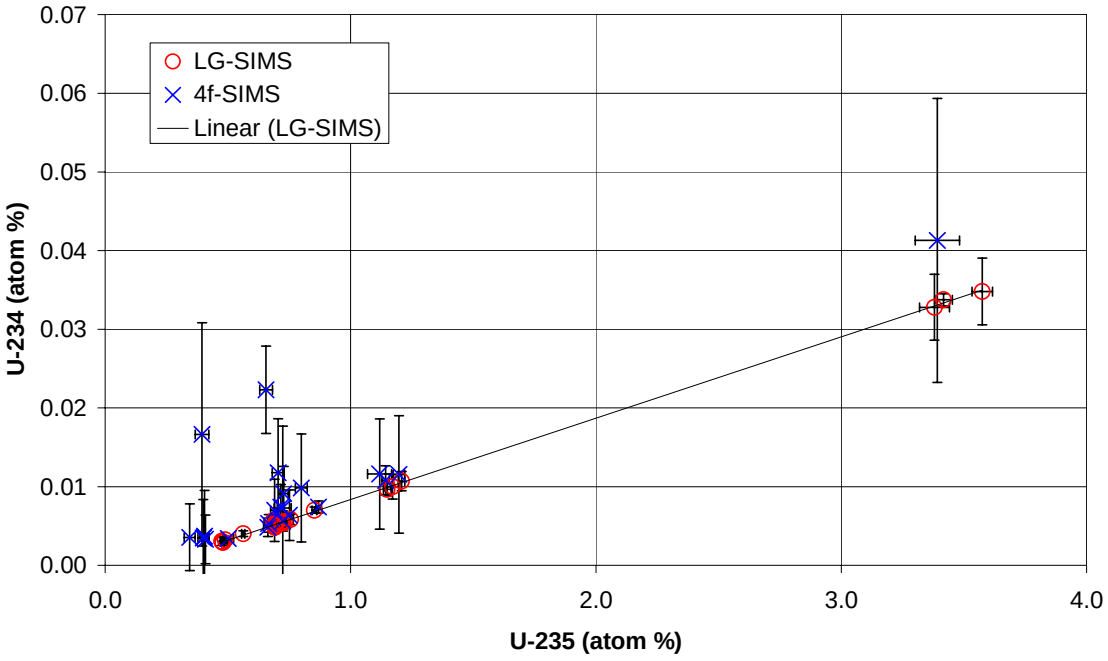

Figure 6. Comparison of 4f and LG-SIMS results for a sample with isobaric interferences.

Figure 6 shows data for both 4f and LG-SIMS for another sample containing abundant, larger particles, but with a high Pb background. Uncertainties are much lower for U-234 from LG-SIMS, associated with

both better counting statistics and reduction in uncertainty associated with interfering species, thus the trend in the data is much better defined by LG-SIMS.

For U-235 abundance, it has not yet been possible to realize an improvement in the accuracy of the data, although the precision is better due to improved counting statistics and better stability of the modern electronics. Experimental artifacts, such as the effect of the particle size, density, and composition on the mass bias correction (up to 1%, relative), have yet to be understood and controlled. In addition, many particles from some samples exhibit changing uranium isotopic ratios during microprobe analysis. This behavior reflects either non-uniform composition within a single (often large) particle (Figure 7), agglomeration of particles during sample preparation, or overlap of the microprobe beam with two separate particles during analysis. Experience has shown that it is advisable to reduce the sample load on an LG-SIMS planchet below the usual level of loading of a planchet for the 4f SIMS, in order to reduce this last artifact.

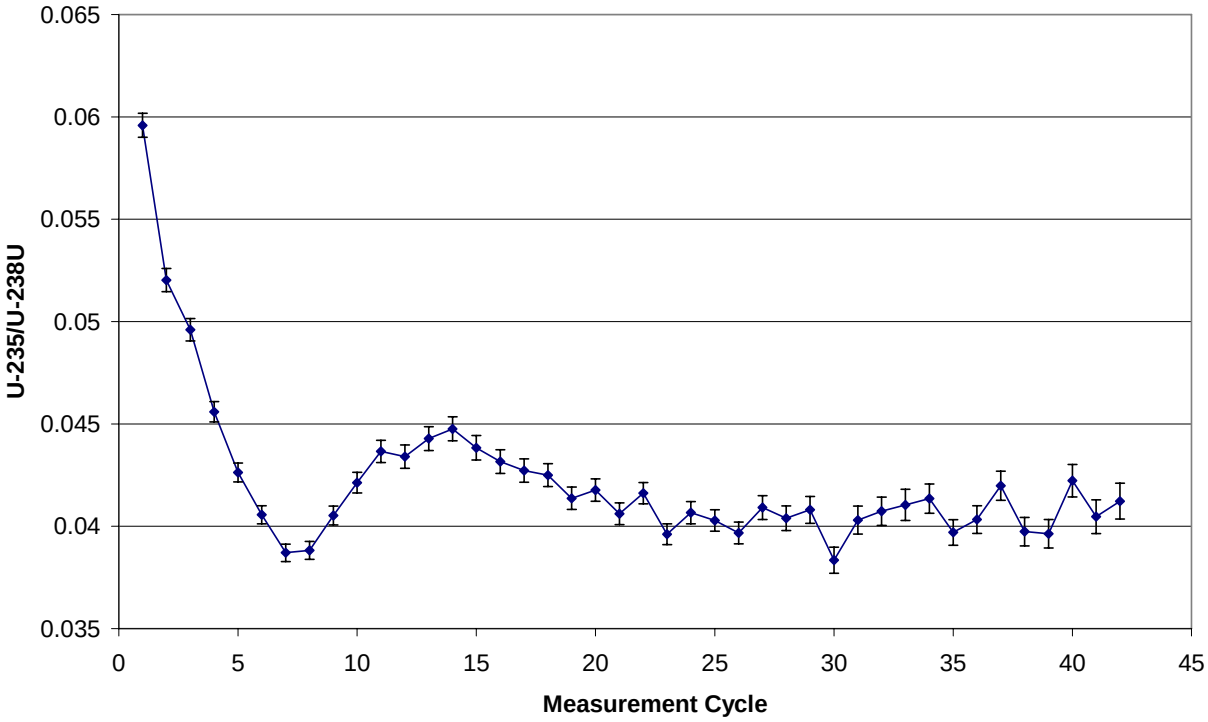

Figure 7. Change in the isotopic composition of a large particle during the course of microprobe measurement.

Measurements for U-236 abundance have been substantially improved due to two factors. First, the correction for the U-235 hydride is based on simultaneously collected data from U-238 hydride. This is an important improvement, since the ratio between U and U-H usually changes by up to a factor of two during the measurement of a single particle. The hydride correction is also typically lower by up to a factor of two for the LG-SIMS compared to the 4f. Second, improvement in counting statistics helps both the U-236 and the hydride determinations. Figure 8 shows data collected over several months for the standard NIST U030a, with an abundance of U-236 of 0.000599 +/- 0.000005 atom percent. These results indicate that such a low abundance of U-236 can be detected on the LG-SIMS in the case of large, clean particles. Although the uncertainties vary depending on the particle characteristics for real samples, preliminary results are encouraging.

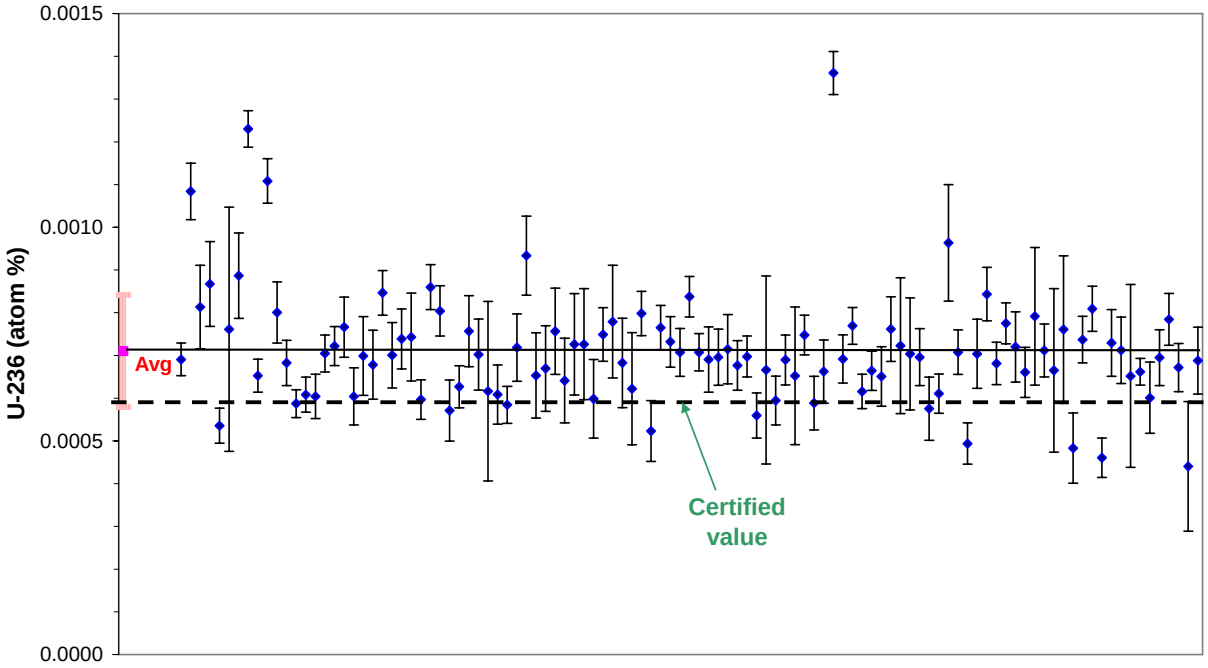

Figure 8: Data for NIST standard U030a (recorded over a period of four months). Low levels of U236 are detectable with a small bias.

4.2 Comparison with FT-TIMS

Figure 9 (below) shows independent data from two swipes taken at the same sampling location, analyzed by LG-SIMS at ESL and by the Fission Track Thermal Ionization Mass Spectrometry (FT-TIMS) technique from the IAEA's Network of Analytical Laboratories. The data are in excellent agreement, although the uncertainties for FT-TIMS are generally smaller. Both data sets are able to discriminate an upward spread of a few of the U-234 values away from a correlation line defined by the majority of the data, outside of experimental uncertainty. LG-SIMS data provide more complete coverage of the depleted end of the isotopic spectrum. These observations have been confirmed by a series of comparisons of data from these laboratories over a range of sample types. These comparisons indicate that ESL's LG-SIMS is capable of meeting the Agency's goal of independently verifying results from the NWAL.

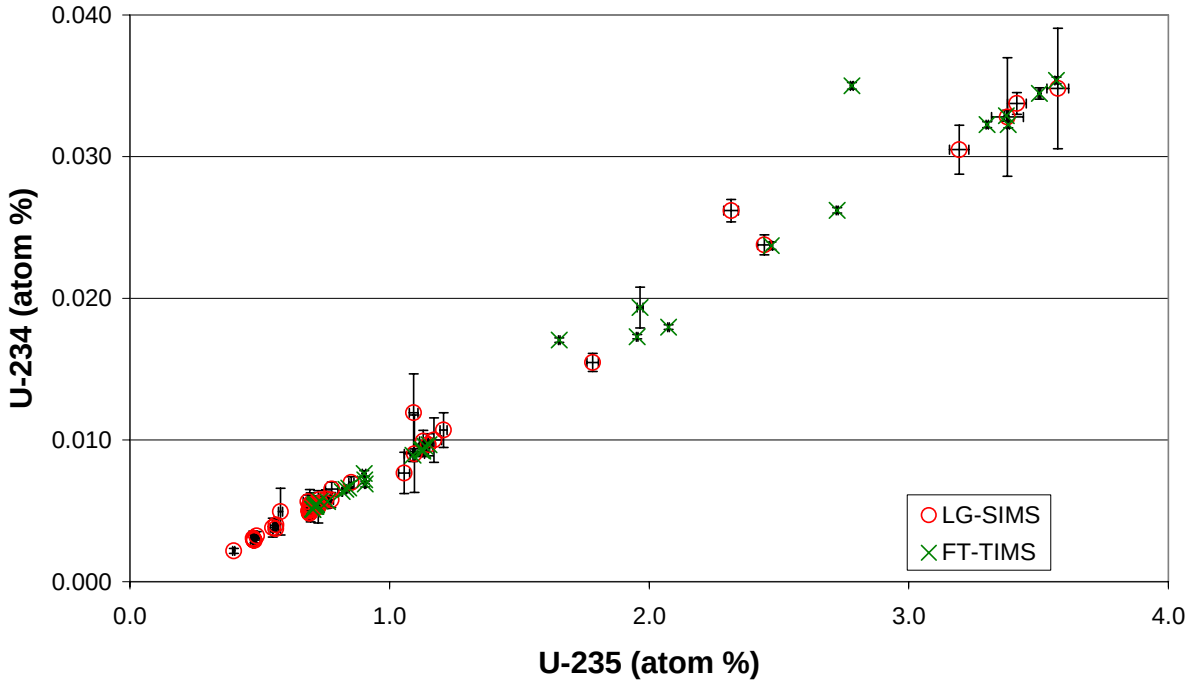

Figure 9. Comparison of LG-SIMS and FT-TIMS data for paired swipes from the same sampling location.

5. Error Estimation

Part of the strategy for rapidly bringing the LG-SIMS into operation was to use error budgets generated during GUM-type uncertainty estimates to understand the factors dominating uncertainty, and use this information to confirm, using various standard materials, that these factors are adequately estimated. A conservative approach is warranted in reporting safeguards data on any new system, but especially one where there is considerable complexity and no long-term experience in the community. Despite this conservative approach, the errors associated with U-234 and U-236 are significantly better than could be generated by conventional SIMS and are small enough to be useful for drawing safeguards interpretations from the data.

Sources of uncertainty that are included in our error propagation (following the "Guide to the Expression of Uncertainty in Measurement") include dead time correction (usually small), detector intercalibration (up to 0.3%, relative uncertainty), mass bias correction (conservatively set at 0.3% per mass unit), counting statistics, a term to reflect variability in the data greater than expected from counting statistics and other known sources of random error, and explicit calculations for changes in the isotopic ratios over time that are greater than expected from counting statistics. U-236 has two additional terms, reflecting the hydride correction, and a term corresponding to a positive bias observed in the U236/238 ratio for standards and natural uranium samples of 1 ppm. Effort is ongoing to identify the source of this bias. An error budget is calculated for every isotope for every particle that is measured, and shows considerable variability in the dominant component, depending on individual particle characteristics.

6. Summary

LG-SIMS is living up to its promise of improving uranium isotopic data for environmental particle analysis. Based on measurement of 16 safeguards samples, the data for U-234 and U-236 are substantially better than those from the Agency's 4f SIMS, and show excellent agreement for data from the same sample by

FT-TIMS from the NWAL, although LG-SIMS is not quite as accurate. LG-SIMS also has the potential advantage of generating larger quantities of data compared to other techniques in use in the NWAL, both moderate quality screening data and more rapid, accurate microprobe data. Improved sensitivity also reveals other sample effects, especially the variability in uranium isotopic composition within single particles. The Agency now has the capability to independently generate high quality results on particles from environmental samples. The future challenges are to continue to improve the accuracy of measurement with LG-SIMS and to incorporate the new types of data that are generated into the process for drawing safeguards conclusions. From the performance of LG-SIMS illustrated above, there are clear benefits to the Agency in taking advantage of additional LG-SIMS capability as it becomes available in the NWAL.

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Feasibility study on the development of uranium reference particles for mass spectrometric analysis in nuclear safeguards

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Abstract:

Recently, an improved method allowing the accurate determination of the isotopic composition of single sub-micrometer sized uranium oxide particles (< 1 μm) has been developed at the Institute for Reference Materials and Measurements (IRMM) in support to nuclear safeguards. This method is based on in-situ particle manipulation using a Scanning Electron Microscope (SEM) and Thermal Ionisation Mass Spectrometry (TIMS) combined with filament carburization and Multiple Ion Counting (MIC) detection. The applicability of the method has been firstly tested on reference uranium particles produced at IRMM, and then applied for the detection of uranium signatures in real life particles collected at a nuclear facility. Moreover, there is an urgent need to provide suitable uranium reference particles, which are not only certified for isotopic abundances but also for the uranium amount contained per single particle. Particularly with the envisaged installation of Large Geometry Secondary Ion Mass Spectrometry (LG-SIMS) instruments, such a particle reference material would be highly appreciated by the International Atomic Energy Agency Network of Analytical Laboratories (IAEA–NWAL) for optimising the overall transmission efficiency in SIMS, TIMS and Laser Ablation Inductively Coupled Mass Spectrometry (LA–ICPMS) instruments used for particle analysis. To face this challenge, IRMM and the Institute for Transuranium Elements (ITU) joined their efforts to develop a method for quantifying uranium in nearly spherical particles by Isotope Dilution (ID)–TIMS. The particles used for this study are monodispersed uranium oxide particles produced at ITU from characterised standard solutions using a vibrating orifice aerosol generator. The goal is to make these certified reference particles available for the validation of measurement procedures, calibration of instruments and as quality control samples for testing the analytical performances of the IAEA–NWAL.

Keywords: Uranium; particle analysis; TIMS; SIMS; isotope dilution; nuclear safeguards

1. Introduction

As part of the response to the Additional Protocol (INFCIRC/540), environmental sampling for safeguards has been adopted by the International Atomic Energy Agency (IAEA) as a strengthened measure for the verification of the absence of undeclared nuclear activities [1]. Particle analysis has been subsequently introduced as a powerful investigative tool for its usefulness in safeguards applications [2]. It is a requirement that the IAEA network of analytical laboratories (NWAL) applies validated measurement methods for safeguards analysis because the conclusions drawn from the measurements might have political and legal consequences on the international scale. Nuclear reference materials (RMs) play a fundamental role in analytical quality assurance as they are widely

used for the calibration of instruments and measurement systems, for method development and validation, or as quality control samples. On the other hand, as the importance of verifying the conformity between the facility declarations and the safeguards obligations increases, new technologies and methods are being implemented that require in parallel elaboration of more complex and appropriate RMs. Despite the efforts among various laboratories to produce materials for safeguard analytical purposes with assigned values that are traceable to the SI, it is a significant drawback that specific kinds of RMs that are matched to a certain type of material are currently not available. In order to supply the scientific community with suitable uranium reference particles, the Institute for Reference Materials and Measurements (IRMM) has developed particles from certified UF_6 reference materials that closely resemble real life particles collected on swipes by safeguards inspectors in nuclear facilities. These test samples, certified for isotopic abundances, were used in the Nuclear Signatures Interlaboratory Measurement Evaluation Programme (NUSIMEP–6) completed in 2008 [3]. Improvements in the preparation and characterisation of such reference uranium oxyfluoride particles have been recently achieved at IRMM [4]. Hence, a NUSIMEP–7 interlaboratory comparison has been launched in March 2011 subsequent to the preparation of new test samples. Nevertheless, there is an increasing demand requiring the availability of uranium reference particle materials, which are certified not only for isotopic abundances but also for the uranium content per particle. Such reference particles would be an essential tool for laboratories carrying out particle analysis in meeting the needs of a quality assurance system and in helping to improve their analytical performances. In particular, such a reference material would be highly appreciated by the IAEA–NWAL for optimising the overall transmission efficiency in Secondary Ion mass Spectrometry (SIMS) – specially with the upcoming installation of Large Geometry SIMS instruments – as well as in Thermal Ionisation Mass Spectrometry (TIMS) and Laser Ablation Inductively Coupled Mass Spectrometry (LA–ICPMS), which are also used for particle analysis. To achieve this goal, IRMM in cooperation with the Institute for Transuranium Elements (ITU) have joined their efforts to develop a reliable analytical method for quantifying uranium in nearly spherical particles by Isotope Dilution Thermal Ionisation Mass Spectrometry (ID–TIMS). The monodispersed uranium oxide particles used in this work were produced at ITU using a commercial aerosol generator (Model 3450 Vibrating Orifice Aerosol Generator, TSI Inc., USA). The experimental set-up for particle generation, which is based on a spray procedure together with the technique of solvent evaporation, has already been described in a previous paper [5]. With this method, particles of well-defined morphology, size and isotopic composition can be obtained. For instance, the mean diameter of the final aerosol generated particles can be fixed by adjusting the concentration of the uranyl nitrate solution in the mixture, which is a substantial advantage for this study. In this work, the procedure used refers partly to the improved method developed at IRMM for the analysis of single uranium reference particles, which has also been applied for measurements of real life particles from a nuclear facility [6, 7]. This method combines particle manipulation using a scanning electron microscope (SEM) with filament carburization and TIMS analysis. First results showed that the applicability of the ID–TIMS method looks very promising for the preparation of uranium oxide reference particles, certified for isotopic abundances and uranium amount content per particle. These particles might be used in the future as certified reference materials for mass spectrometric analysis in nuclear safeguards.

2. Experimental

2.1. Materials

The uranium oxide particles used in this work were prepared starting from an aqueous alcoholic solution of uranyl nitrate obtained by dissolution of the certified reference powder material CRM U100 (10 % enriched in ^{235}U) from New Brunswick Laboratory (Argonne, USA) with 8 mol L^{-1} HNO_3 (Merck, Germany). The resulting solution was firstly adjusted to the concentration of ca. 50 mg U g^{-1} in 1 M HNO_3. The final solution injected into the aerosol chamber (ca. 70 mg U L^{-1} in a 50:50 mixture of isopropanol and water) was adjusted accordingly to generate particles of ca. 1 µm. After calcination and release from the cooler, particles are typically collected for 1 h on a Nucleopore filter and stored under vacuum. In order to perform SEM energy dispersive X–ray (EDX) analysis, particles were transferred on a high purity carbon planchet (Grade A Carbon Planchets No. 17680, 25 mm Ø, E.F. Fullam, Inc., USA) using a vacuum impactor system [8].

For IDMS analysis, a ^{233}U spike isotopic reference material IRMM–058 solution with a uranium concentration of 2.20 pg U μL^{-1} (in HNO_3) was used. It was transferred from the original flame-sealed quartz ampoule into a pre-cleaned teflon bottle. In addition, a concentrated solution of 8 mol L^{-1} HNO_3 was prepared by dilution of 65 % ultrapure nitric acid (Merck, Germany) with deionized Milli-Q water. This strong acid solution was used for the second set of measurements (see next paragraph) in order to ensure the completeness of the particle dissolution. All reagents used in this work were of analytical grade. Teflon bottles used to store the solutions were previously washed according to a cleaning procedure specific for ultratrace analysis.

2.2. Instrumentation

2.2.1. Scanning electron microscopy

The particle transfer was performed within the vacuum chamber of a tungsten-filament FEI Quanta 200 3D scanning electron microscope (SEM) using a special holder to carry the planchet and the TIMS filaments [6]. Either a single or a fixed number (max. 6) of U–oxide particles were transferred on pre-degassed and carburized single rhenium filaments using a micromanipulator MM3 (Kleindiek Nanotechnik, Germany) equipped with a solid tungsten needle (Model ST–20–0.5). Prior to manipulation, the elemental composition of the U–oxide particle was checked by EDX and the particle size estimated by means of SEM images (Fig. 1). The uncertainty of the particle diameter is estimated to be about 5%, which causes the uncertainty of the derived particle volume to be at the order of about 15%. An example of the particle loading process is shown in Fig. 2.

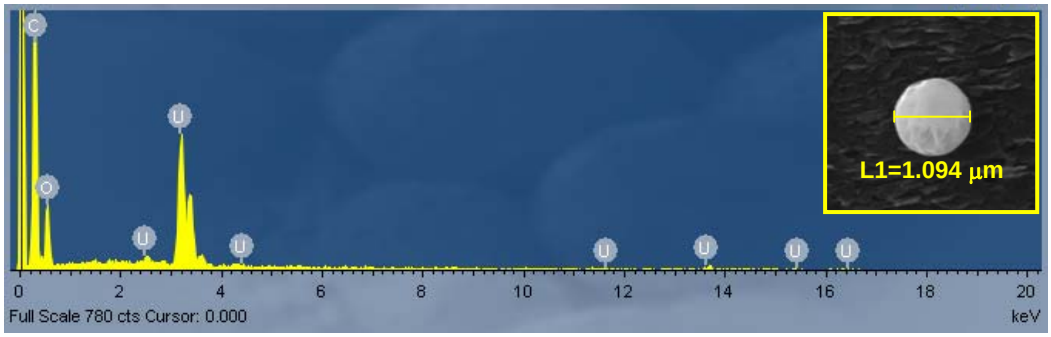

Figure 1: Uranium oxide particle of about 1 µm identified by an EDX spectrum.

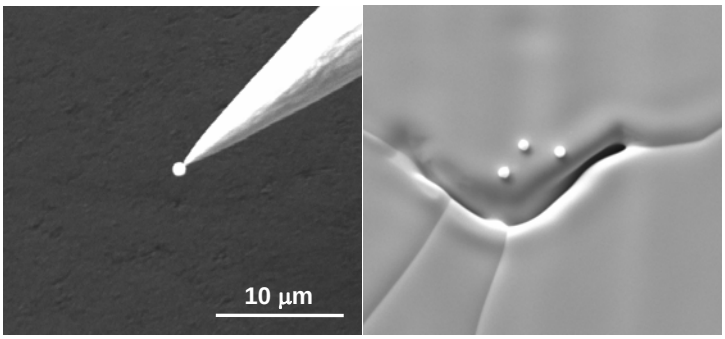

Figure 2: Scanning electron microscope images of U100 particles transferred on a carburized rhenium filament using a micromanipulator.

2.2.2. Thermal ionisation mass spectrometry

Isotope ratio measurements were carried out using a TRITON TIMS (Thermo Fisher Scientific, Bremen, Germany) equipped with a multiple ion counting system (MIC) and a secondary electron multiplier detector. In this work, the secondary electron multiplier was operated in the ion counting mode to enable the measurement of very low ion signals, and the isotopes of interest (^{233}U and ^{238}U)

were collected with a peak jumping measurement procedure. Filament carburization was performed using pure benzene gas as a carbon provider for enhancing the overall ionisation efficiency of uranium in the TIMS ion source. For IDMS analysis, each sample filament was spiked with a known amount of IRMM–058 using a calibrated monocanal micropipette (Eppendorf, Germany). Two different sets of measurements were performed. The first set included samples of only particle(s) plus spike while the second set additionally contained one or several μl–drops of 8 mol L^{-1} HNO_3 added directly on the filament before spiking in order to further ensure the dissolution of the uranium oxide particles. The amount of spike was increased from 1 to 4 μl with increasing number of particles (1–6) loaded on the filament surface. In order to estimate the U blank value, filaments loaded with different amounts of spike and spike plus 8 mol L^{-1} HNO_3 were also measured. Each sample filament was mounted together with a modified side filament on a 21 position turret and introduced into the TIMS ion source. Prior to analysis, prebaking of these modified side filaments was performed according to a specific procedure (1750 °C for 1 h per position) found to be helpful for decreasing the background levels by a factor of 5–10 [6].

3. Results and discussion

Experimental data obtained from the two measurement sets are reported jointly in Fig. 3 as the total mass of uranium measured by ID–TIMS versus the total volume. Each data point displayed results from the measurement of one filament. The total volume represents the accumulated volume for all particles loaded on the same filament and estimated from the particle diameters measured using the SEM images. The uncertainty of the results is mainly due to the uncertainty in pipetting the spike solution, the estimation of the particle volume (calculated from the estimated particle diameters) and the measurement uncertainty. As one can see, the results show no difference in the two approaches used (with and without additional HNO_3 8 mol L^{-1}), therefore the nitric acid of the IRMM-058 spike solution (< 5 mol L^{-1}) seems to be sufficient for dissolving most of the particles. No significant change in slope was observed by applying the HNO_3 blank correction to the data. An estimation of the average uranium mass concentration within the uranium oxide particle can be calculated from the slope of the regression line reported in Fig. 3, and the value obtained is 4.54 ± 0.78 g cm^{-3}. Depending on the particle stoichiometry, the density can be derived from such data. For instance, if the assumed stoichiometry for the uranium oxide particle is U_3O_8, the corresponding density given by the product of (4.54×100/84.8) is 5.35 ± 0.92 g cm^{-3}; it would be 5.46 ± 0.94 g cm^{-3} (4.54×100/83.2) if the assumed stoichiometry is UO_3. Both these values are relatively far from the literature values of the densities for U_3O_8 and UO_3, which are 8.38 g cm^{-3} and 7.0 g cm^{-3}, respectively [9]. This means that particles are probably composed of microspheres of highly defected material with a lower density than that expected for a pure crystalline U-oxide form.

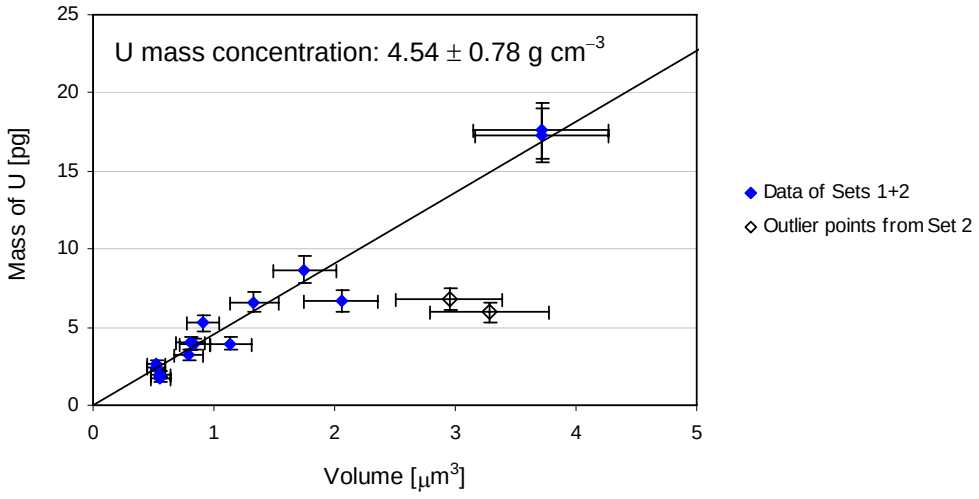

Figure 3: The graph shows the relationship between the total mass of uranium measured by ID–TIMS versus the total volume (accumulated).

In Fig. 4a, the total mass of uranium measured by ID–TIMS is plotted versus the number of particles. For comparison, the graph obtained by Ranebo *et al.* [10] from ID-TIMS measurements of three different U020–A (2 % enriched in ^{235}U) samples containing 30, 43 and 118 particles, respectively, is reported in Fig. 4b. As one can see, there is a close agreement between the experimental value of the slope obtained in this study (2.80 ± 0.28 pg) and the literature value (2.59 ± 0.12 pg). But this finding has to be further confirmed because the U–oxide particles used by Ranebo *et al.* were prepared with a different production setting and they were additionally suspended in isopropanol and dissolved in 5 M HNO_3 prior to the ID–TIMS analysis. Ranebo *et al.* reported an average process blank of 57 pg as it can be seen from the y-intercept in Fig 4b. This uranium background component was ascribed to the isopropanol from the suspension. In this work, a U blank value in the range of only 0.026–0.14 pg was achieved by loading the particles directly on the filament surface without the addition of other chemicals. However, it is not clear whether the particle(s) loaded on the TIMS filament are completely dissolved and homogeneously mixed with the spike. A good indicator is given by the monitoring of the $n(^{238}U)/n(^{233}U)$ ratio during the course of the TIMS measurement. For instance, significant sudden increases of the $n(^{238}U)/n(^{233}U)$ ratio during two measurement runs might explain the outlier points of the second set represented by the open circles in Fig. 3. But for the rest of the data it can be assumed that the particles are rather well dissolved by the nitric acid because of the rather small variations (few percents) of the measured $n(^{238}U)/n(^{233}U)$ ratio during the TIMS analysis. So far the critical step for the method validation is related to the investigated sample, which is not composed of perfectly spherical particles. Moreover, the particle size distribution observed by SEM was found to be in a relatively broad range of 0.7–1.3 μm. This was ascribed to the particle generation process in which the U-oxide particles might result from the coagulation of multiple droplets [5]. Also the presence of "voids" suggesting that the material was not homogeneously distributed within the particles could be observed, thus affecting the U mass concentration that might vary from one particle to another.

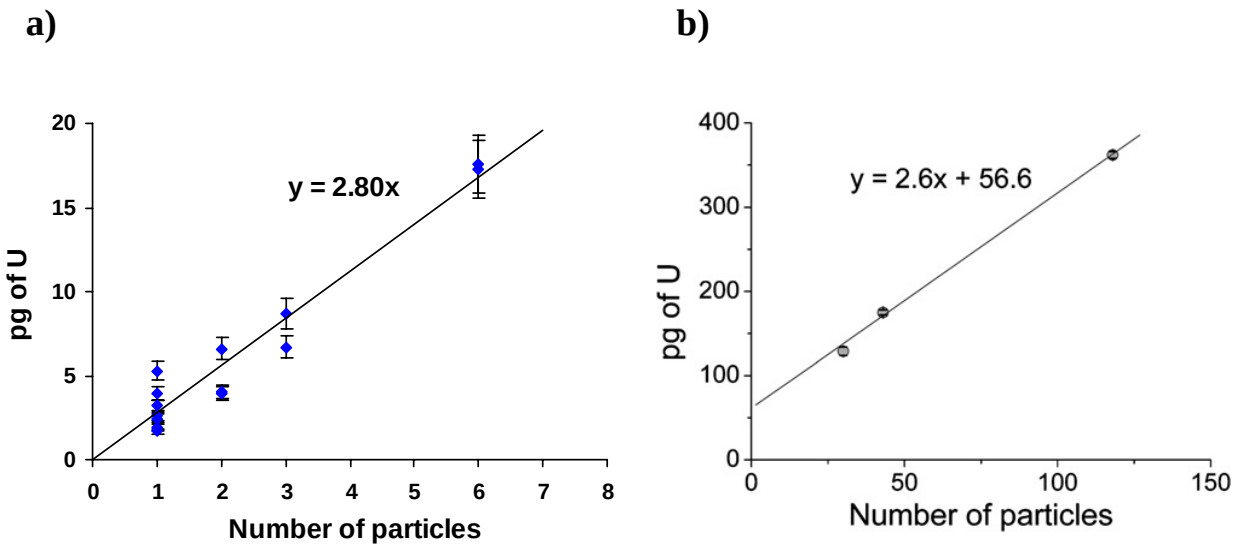

Figure 4: Relationship between the total mass of uranium measured by ID–TIMS versus the number of particles obtained a) in this work and b) by Ranebo *et al.* [10]

4. Conclusions

Although the measurements performed on the U100 uranium oxide particles are only a starting point, this feasibility study showed that the ID–TIMS method developed at IRMM is well suited for quantifying the amount of uranium contained per particle. The main limitations of the method can be ascribed to the large uncertainty in estimating the particle size (diameter) from SEM images and to the completeness of the particle dissolution using the spike either with or without 8 mol L^{-1} HNO_3 on the TIMS filament. In particular, it is important to have the production process of representative uranium particles with well-defined size under control in order to be used by the mass spectrometric community for determining respective ion yields. For this reason, the applicability of the method needs to be

further tested via experiments carried out on more uranium particles from new batches. For a future reference material certified for the U amount content, the settings in the aerosol generator have to be fixed in order to ensure a reproducible monodispersed uranium particle production. Additionally, for a better understanding of the difference observed for the estimated average density in comparison to the literature values, we also need to take into account additional parameters, e.g. particle morphology, stoichiometry and processing temperature during production and calcination, which might affect the material's density. The overall conclusion from this feasibility study is that the future production of a uranium particle reference material certified for isotopic abundances and amount content per particle in support to the IAEA-NWAL looks very promising.

5. Acknowledgements

The authors would like to acknowledge Mr H. Thiele from ITU (Karlsruhe, Germany) for its help with the SEM–EDX analysis prior to sample shipment to IRMM.

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Environmental TBP and DBP emissions as an indicator for unreported reprocessing or uranium refining and conversion activities

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Abstract:

In 2005, the Department of Safeguards of the IAEA started the "Novel Technologies Project", looking for novel techniques to detect unreported nuclear activities. Aims of this project are on the one hand to build up a database collecting all information about possible indicators and signatures suitable for nuclear safeguards. On the other hand, novel technologies are needed that inspectors could apply to detect such indicators or signatures possibly hinting at unreported activities or materials [1].

One possibility might be the detection of dibutyl phosphate (DBP) in liquid emissions of nuclear plants. It could serve as an indicator for both uranium enrichment and an indicator of reprocessing. In this paper the DBP source term and detection techniques are discussed and also other legitimate application of DBP are introduced.

Keywords: Environmental sampling, tributylphosphate, dibutylphosphate, novel technologies, reprocessing, uranium conversion

1. Uses of TBP and DBP in nuclear industry

Tri-n-butylphosphate (TBP) is used in large quantities in the nuclear industry and of proliferation concern because it is a crucial non-nuclear material for plutonium separation and for uranium refining. In 2002, North Korea received a shipment of 30 tonnes TBP from China. In 2006 and 2007, the Indian Department of Atomic Energy procured about 250 tonnes of TBP from Germany and Russia.

TBP is used for the solvent extraction process to concentrate and purify nuclear-grade uranium in theyellow cake (U_3O_8). Uranium is further refined at a conversion facility. After dissolution of the yellow cake in nitric acid the solution of uranium nitrate is treated by a solvent extraction processusing TBP dissolved in kerosene or dodecane. The product of this purification step is UO_3that is further reduced to UO_2 and then fluorinated first to UF_4 and then to UF_6.

TBP is used in various reprocessing schemes for the separation of plutonium from uranium fuel and for separation of uranium from thorium fuel. In particular TBP is the solvent for the liquid-liquid-extraction of PUREX for the recovery of uranium and plutonium from spent nuclear fuel discharged from Light Water Reactors. Commonly in use is a mixture of 30% TBP and 70% kerosene or, more rarely, of dodecane. This TBP 30 meets all the requirements of nuclear fuel reprocessing: Good physical properties and chemical stability, suitable extraction powers and decontamination from fission products, very low corrosion and solvent degradation effects. Since the irradiated fuel is dissolved in

nitric acid, the nitrates of uranium and plutonium are formed. These nitrates form complexes withTBP,hence staying in organic phase while the other fission products stay in the aqueous phase. The uranium and plutonium can be extracted in this way.Although there are other reprocessing processes available PUREX is the international standard for spent fuel reprocessing.

Dibutylphospate (DBP) is a by-product of nuclear process involving TBP since it is generated by radiolysis from TBP.

2. Radiolysis

TBP is used because of its high chemical stability. It is almost insoluble in water, non-combustible and does not react with the nitric acid that is used to dissolve the irradiated fuel [2]. Nevertheless it can be degraded by radiolysis. The main product of this radiolysis is DBP and further radiolysis yields monobutyl phosphate (MBP). Further degradation products are alcohols, ketones, nitrates, nitrites and various phosphates. The DBP forming reaction is shown in figure 1. Since TBP and DBP are non-radioactive after use they are recycled within the plant. Small amounts remain in aqueous waste streams that are disposed into the rivers or ocean near reprocessing plants. These traces can be measured in water samples. High concentrations of DBP near a nuclear plant can be a possible evidence of declared or undeclared reprocessing activities. High concentrations of TBP could indicate both reprocessing as well as preparatory steps for uranium enrichment.

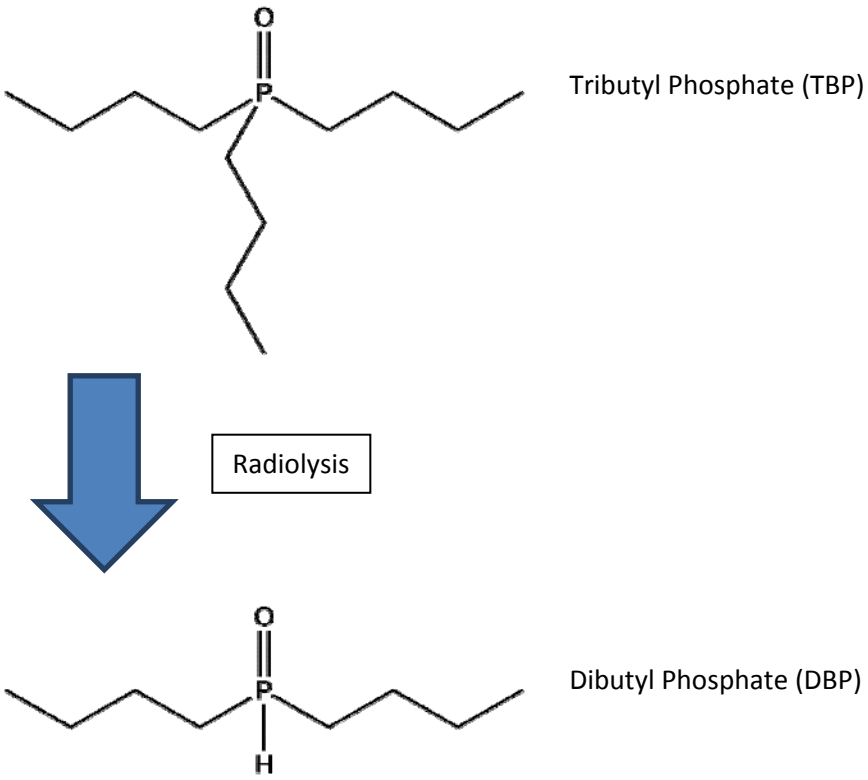

Fig.1: Simplified reaction scheme for the radiolysis of TBP

Since it is impossible to find exact values for the amount of DBP released in liquid effluents, an indirect way must be chosen to reconstruct them. There are several publications on the radiolysis of TBP during PUREX. This is why the yield of DBP in this process could be estimated quite well. Table 1 shows DBP yields from TBP radiolysis [4].

Dose, rad[1]	Energy absorption, Watt hr / litre	Yield, g/watt hr
10^4	0.027	1.05
10^5	0.27	0.27
10^6	2.7	0.17
10^7	27	0.17
10^8	270	0.17

Table 1: DBP yield by radiolysis of TBP [4]

For measuring the data presented in table 1, a mixture of 30% TBP with iso-octane has been used. The conditions are therefore comparable to the situation in a reprocessing plant. The table shows the yield of DBP per energy deposited by radioactive decayin the TBP component of the mixture.As can be seen in Table 1, a dose increase results in a decrease of the yield. At high radiation levels, the yields are independent of the dose. The largest yield occurs at the low end of the investigated irradiation levels[5].Based on the fact that the amount of radiation received in most processing applications is less than 0.1 watt-hour per litre[5]a maximum concentration in the unit gram per litre can be assessed. Assuming an energy absorption that is just a quarter of the stated maximum (0.025 Wh/l), the resulting yield would be about 1 g/Wh according to the first row in Table 1. Hence, the concentration of DBP in TBP would be 25 mg/l. This implies that about 2.5% of TBP would be degraded to DBP.[2]Each reprocessing activity would increase the fraction of DBP. To prevent it to build up too far, the solvent is periodically washed with an alkaline solution.

The further degradation products due to radiolysis are MBP and phosphoric acid (H_3PO_4). The ratios of thethree products TBP, DBP and H_3PO_4are4:2:1 [3]. However, due to various recycling and cleaning processes before any material ends up in a release to the environment, these ratios cannot be expected to be found in the releases. Nevertheless, it can be assumed that the DBP to TBP ratio is significantly higher than by normal decomposition processes of TBP without radiolysis. Hence, the high DBP to TBP ratio is a very specific indicator for nuclear reprocessing.

3. Reported releases to the environment

In table 2 the amount of effluents that a reprocessing plant emits per year can be seen [5]. The examples given are the data for the reprocessing plant at La Hague in France from 2003 to 2007.

year	TBP [kg]	Total phosphor [kg]
2003	1500	2600
2004	2100	2600
2005	2320	2670
2006	2560	2770
2007	1680	710

Table 2: Liquid effluents discharges of TBP and phosphate from the La Hague reprocessing plant [5].

Figure 2 shows the amounts of discharges of TBP and total phosphor. The amount of elemental phosphorreported as “total phosphor” includes this element in all possible chemical forms other than TBP, in particular DBP, MBP, phosphoric acid and other chemicals containing phosphor.
There are also release data available from British Nuclear Fuels. BNF discharged even more TBP into the water, about 30 tonnes per year from 1995 to 1997 [7]. About two thirds come from Sellafield where used nuclear fuel is reprocessed. About one third is from Springfields where uranium is produced in a centrifuge enrichment plant. The numbers can be seen in table 3.

[1] 1 rad = 1 J/kg = 2.78 *10^{-6}Wh/kg
[2] The densities are 0.98 g/cm^3 for TBP and 1.058 gcm^3 for DBP.

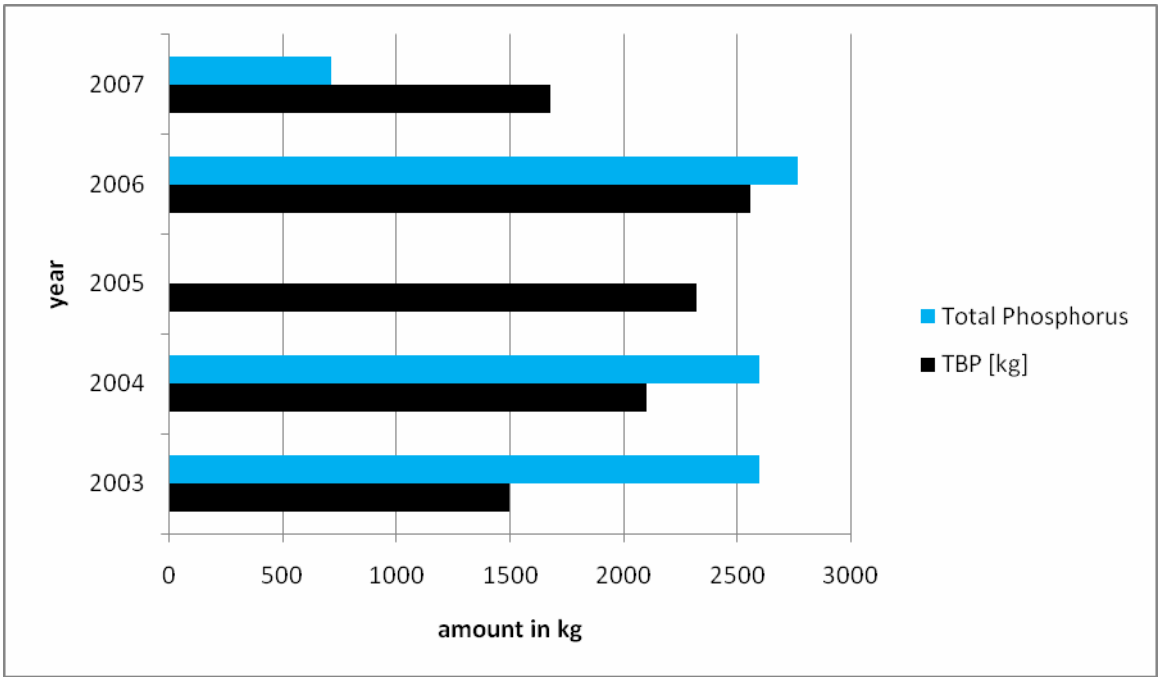

Fig. 2: Comparison of TBP discharges and total phosphordischarges.

In practice, as assumed before, mostTBP is not discharged together with the degradation products dibutyl phosphate, monobutyl phosphate and other phosphates. Before the aqueous waste stream containing DBP is released, its TBP content is washed out with a clean solvent. As a result, the amount of discharged DBP cannot be calculated. From total phosphor measurements and based on the fact that DBP is produced in considerably higher yields than any other degradation product, it can be estimated that the amount of DBP released to the environment can well be in the order of 10% of the TBP emissions, hence about 150 to 250 kg DBP from La Hague in the years 2003-2007.

Year	1995	1996	1997
TBP in tonnes	33	34,1	**31,5**
		Sellafield	21
		Springfields	10,5
			31,5

Table 3: Liquid effluents discharges of the BNFL [7]

4. Environmental concentrations

Concerning environmental concentrations, La Hague in France measures the concentration of TBP and total phosphates 2 hours after emission. According to environmental protection standards the maximum value allowed for total phosphate is 1 µg/l (on average) [6].Actually reported are levels of less than 0.5 µg/l for TBP and less than 0.7 µg/l for total phosphormeasured in seawater near La Hague [5]. The BNFL did not give concentration values in its annual report.

5. Measurementtechnologies

In the laboratory, standard technologies for determining low concentrations of TBP and DBP are high-pressure liquid chromatography (HPLC) and for total phosphorinductively coupled plasma mass spectroscopy (ICP-MS). Areva states detection limits for the effluents as 5 mg/l for total phosphorby ICP-MS and 3 mg/l for TBP by HPLC. DPB is not separately measured [5].

In order to facilitate in-field measurements, several systems for detection of TBP and DBP in the aqueous phase are currently under development. These should be portable and capable to generate a fast result. This would enable an inspector to decide prompt on further measures in the case of a suspicious detection.

- The use of ytterbium as a sensor to detect spectrophotometricallyphosphate anions is studied at the Shanxi University in Taiyaun, China. The detection limit is 10^{-5} mol per litre for TBP.
- The use of colorimetric sensor array is developed by Chemsensing in Illinois, USA. Chemically responsive dyes are used for the molecular recognition of organic molecules. The detection limit is 10^{-6} mol per litre for TBP.
- A miniaturized and portable capillary electrophoresis system (MinCE) hasbeendeveloped at the Instrumental Analysis Institute in Karlsruhe, Germany (IFIA, FZK). The separation of a substance is achieved by its mobility in an electric field. This method is capable to determine TBP, DBP and MBP in aqueous solutions. The detection limit is10^{-6} mol per litre[8].

As shown in the previous section, all approaches for in-field measurements have in common that the lower end of the working range is not sensitive enough for the background level in environmental samples. These are usually lower than this sensitivity and would therefore require samples to be analyzed in the laboratory. However, for cases with extraordinary high concentrations, capillary electrophoresis and colorimetry are most promising with a sensitivity of 10^{-6} mol per litre in laboratory tests and working in concentrated nitrate media [9]. This equals a concentration of 270 µg per litre for TBP and210µg per litre for DBP.[3]This performance is better than the standard laboratory analysis for effluents as described above. However, compared to the maximum value of 1 µg total phosphate which is allowed in French waters, this value is still very high.

6. Interferences

Another factor that should be taken into consideration is that both TBP and DBP have many industrial applications. TBP is frequently used as a solvent for extraction and purification of rare earth metals from ores. Other uses are as a plasticizers, catalyst antioxidants, antifoaming agents and corrosion inhibitors. It can be found as a component to hydraulic fluids in aircrafts, in adhesives, synthetic resins and inks. DPB is often used in the textile industry as an antistatic agent. Furthermore it can be used for dry cleaning to increase the efficiency and the corrosion protection. And finally it is used for polyurethane products [10]. All of these can be legitimate sources of DBP and are therefore an interfering factor for the measurement of DBP to detect reprocessing activities. For example in the USA between 10,000 and 500,000 pounds were produced in 2002 and the years before since 1986 [11].

7. Conclusions

The following conclusions can be drawn. Since unexpectedly high concentrations of TBP and DBP can be an indicator for plutonium production or for uranium refinement and conversion, their analysis in environmental samples might be used as a novel approach for nuclear safeguards to detect undeclared nuclear activities. At present, TBP is measured regularly in the analytical laboratories of reprocessing plants for the purpose of environmental monitoring. This data is therefore readily available and can be used for determining normal background levels.

New in-situ measurements techniques have recently been developed for this purpose that could be applied by inspectors in the field. Two key advantages would be (1) a fast availability of data by

[3] The molecular weights are 266 g/mol for TBP and 210 g/mol for DBP.

handheld measurement devices and (2) an additional way of getting important safeguards information from outside of the site by measurements of TBP/DBP concentrations in wastewater discharges.

The measurement technology must be able to measure very low concentrations in water samples at environmental background concentrations down to about 0.1 µg/l. For direct sampling of effluents a detection limit of about 1 mg/l may be sufficient. Currently available measurement technologies for in-field analysis reach detection limits of approximately 0.3 mg/l for TBP and 0.2 mg/l for DBP. The real requirements, typical environmental concentrations and the measurement performance require further studies to determine the applicability of environmental DBP and TBP detection as a novel tool for nuclear safeguards.

Furthermore because of the various commercial uses of TBP and DBP it is not easy to distinguish relevant detections from background concentrations caused by legitimate sources. However, a high DBP to TBP ratio is a very reliable signature specific for nuclear reprocessing.

8. Acknowledgement

This study has been made possible by the funds received from the DSF (Deutsche StiftungFriedensforschung). I thank Paul Kaiser for editorial support.

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15 NDA I - Neutron detectors

^{3}He Replacement for Nuclear Safeguards Applications - an Integrated Test Program to Compare Alternative Neutron Detectors

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Abstract:

During the past several years, the demand for ^{3}He gas has far exceeded the gas supply. This shortage of ^{3}He gas is projected to continue into the foreseeable future. There is a need for alternative neutron detectors that do not require ^{3}He gas. For more than four decades, neutron detection has played a fundamental role in the safeguarding and control of nuclear materials at production facilities, fabrication plants and storage sites worldwide. Neutron measurements for safeguards applications have requirements that are unique to the quantitative assay of special nuclear materials. These neutron systems measure the neutron multiplicity distributions from each spontaneous fission and/or induced fission event. The neutron time correlation counting requires that two or more neutrons from a single fission event be detected. The doubles and triples neutron counting rate depends on the detector efficiency to the 2nd and 3rd power, respectively, so low efficiency systems will not work for the coincidence measurements, and any detector instabilities are greatly amplified. In the current test program, we will measure the alternative detector properties including efficiency, die-away time, multiplicity precision, gamma sensitivity, dead-time, and we will also consider the detector properties that would allow commercial production to safeguards scale assay systems. This last step needs to be accomplished before the proposed technologies can reduce the demand on ^{3}He gas in the safeguards world. This paper will present the methodology that includes MCNPX simulations for comparing divergent detector types such as ^{10}B lined proportional counters with ^{3}He gas based systems where the performance metrics focus on safeguards applications.

Keywords: nuclear safeguards, NDA instrumentation, neutron detectors, gas proportional counters

1. Introduction

During the past several years, the demand for ^{3}He gas has far exceeded the gas supply. This shortage of ^{3}He gas is projected to continue into the foreseeable future. There is a need for alternative neutron detectors that do not require ^{3}He gas. For many decades, neutron detection has played a fundamental role in the safeguarding and control of nuclear materials at production facilities, fabrication plants and storage sites worldwide. Neutron measurements for safeguards applications have requirements that are

unique to the quantitative assay of special nuclear materials. These neutron systems measure the neutron multiplicity distributions from each spontaneous fission and/or induced fission event. The neutron multiplicity time correlation counting requires that three or more neutrons from a single fission event be detected. This triples neutron counting rate depends on the detector efficiency to the 3rd power, so low efficiency systems will not work for the multiplicity measurements, and any instabilities are greatly amplified.

In the Los Alamos National Laboratory test program, we will measure the detector properties listed in the next section, and we will also consider the detector properties that would allow commercial production to safeguards scale assay systems. This last step needs to be accomplished before the proposed technologies can reduce the demand on ^{3}He gas in the safeguards world.

For most applications related to nuclear security, the primary goal of the neutron measurement is to have good sensitivity for neutron sources at a distance and to have minimal interference from gamma- ray activity and background noise. The identification of the neutron source together with a good lower limit of detection are the focus of the measurement. On the other hand, the primary task for neutron measurements in nuclear safeguards and non-proliferation is to determine the mass of the special nuclear material (SNM) to verify that material has not been diverted. In most cases, the accuracy of the measurements have to be better than 1-2% to meet IAEA international obligations under agreements such as the Non-Proliferation Treaty (NPT). Large scale nuclear plants such as mixed oxide (MOX) fabrication plants process tons of plutonium per year, and the high accuracy of the measurement systems used to verify the plutonium is critical. The ^{3}He based neutron NDA systems have been under development, implementation, and continuous improvement over a four decade period. The result of this development has resulted in a variety of ^{3}He based NDA systems that can provide a precision of 0.1% and an accuracy of 0.3% for plutonium inventory sample measurements in actual plant environments.

The primary purpose of the Los Alamos test program is to evaluate neutron detectors for potential replacement of 3He tubes with an emphasis on the parameter space that is important in nuclear safeguards applications. The parameters that will be measured include:

- **Efficiency** - The total system efficiency for coincidence counting needs to be similar or better than the ^{3}He based systems.
- **Gamma Discrimination** – Safeguards systems must operate normally for dose levels up to 1-10 R/h on the detector face to accurately measure bulk Pu samples containing ^{241}Am. The gamma/neutron ratio should be better than 10e-8.
- **Stability** – The precision and stability for the ^{3}He based systems have been demonstrated to be better than 0.1% for in-plant measurements; however, for simple neutron monitoring, less stability can be acceptable. Stability and reliability are critical for accurate results when instruments must operate continuously in unattended mode.
- **Dead-Time** – Neutron counting at rates up to 1-5 MHz with accurate dead-time corrections are needed for a multi-tube system. Each amplifier module should be capable of processing up to 0.3 MHz.
- **Die-away time** – Neutron die-away time (or lifetime) should be less than 50 microseconds. Because of the neutron coincidence counting application in safeguards, the neutron die-away time of the system is important to reduce the gate length and the accidental counts in the background gate.
- **Detector size** – For most safeguards applications, the high efficiency for measuring fast neutrons from the sample has to be obtained in a relatively small foot print. The high efficiency of ^{3}He permits compact design. The optimum use of hydrogenous neutron moderator needs to be integrated into the design.

- **Scalability** – For many applications, the detector geometry has to surround the sample to provide the high efficiency and to be independent of the sample's shape and distribution. The sample volumes vary from vials to crates resulting in detector volumes that are large.
- **Safety** – The in-plant installed systems have criticality, fire, and seismic safety requirements that are stringent for uranium and plutonium processing plants.
- **Survivability** – Neutron detectors that are used in safeguards applications must be able to function for long periods (years) under the continuous irradiation of both neutrons and gamma-rays without a degradation in performance.

2. Test Methodology

The test program will include a variety of detector types, shapes and sizes that were determined by the fabricators of the systems and not by the test program. The application of ^{3}He detectors for safeguards and nonproliferation has historically included a wide array of shapes, sizes, and efficiencies. The current safeguards test program needs to have a method to normalize the different geometries to be compared with each other and to a ^{3}He based system. This will be accomplished by using the Monte Carlo Neutron and Photon ext (MCNPX) simulation code [1] that has had benchmark measurements for a ^{3}He based system located in the testing laboratory. The ratio of the measured response from the alternative replacement detector will be compared with a virtual "reference" ^{3}He detector of the same size and face area using MCNPX calculations. The MCNPX calculations will be benchmarked with measurements using an actual ^{3}He slab detector in the same position as for the test detectors. All of the test detectors will be configured in a slab geometry with optimum HDPE moderator surrounding the detector for ^{240}Pu fission spectra neutron detection with maximum efficiency.

The ^{3}He slab detectors come in a large variety of sizes, moderator thickness, number of ^{3}He tubes, and ^{3}He gas pressure. To be able to compare the new detector options with a ^{3}He based system, we will define a ^{3}He tube "reference case". The ^{3}He reference has been selected to match a typical ^{3}He detector slab with one row of tubes that is optimized for efficiency and cost. For several decades, the safeguards choice has been 4 atm ^{3}He tubes (1" diameter) with approximately a 5 cm tube spacing pitch There is 2-4 cm of HDPE moderator on both ends of the ^{3}He tube array. The centerline for the tubes is located about 3.8 cm from the front face of the HPPE with some variation in depth depending on the average neutron source energy spectrum. For our present test program, we are using three different neutron source spectra (bare ^{252}Cf, shielded ^{252}Cf, and ^{240}Pu). For safeguards relevance, the detector moderator will be optimized for the ^{240}Pu spectra, and not modified for the other neutron sources.

Our ^{3}He reference detector for comparison with the alternative detectors will have the same face width, active height, and depth as the new detector that is under test, and the efficiency of the ^{3}He system will be determined by MCNPX simulations plus the benchmark measurements. The ^{3}He reference normalization ratio to the new detectors will provide the efficiency comparison between the various alternative detectors in the test program. Our initial tests have focused on ^{10}B lined proportional counters, and future tests could include ^{6}Li, and ^{10}B doped scintillation detectors.

When using ^{3}He detector tubes, the high voltage (HV) bias on the anode wire is typically set at approximately 40 volts above the "knee" in the plateau curve. This makes the efficiency relatively insensitive to small variations in the HV supply. However, for applications in high gamma-ray areas, the HV is lowered so that the operating voltage is about 5% below the plateau level. Under these conditions,

a 30 cm long ^{3}He tube with an optimized preamplifier can count neutrons without gamma interference up to dose levels of more than 20 R/h.

3. Efficiency Considerations

The efficiency of neutron detectors is a function of the neutron moderator design and the thermal-neutron capture reaction with isotopes such as ^{3}He, ^{10}B, and ^{6}Li. Figure 1 illustrates the reaction cross-sections for the most obvious replacement isotopes. We see that ^{3}He has the largest cross section but ^{10}B is only 30% lower. The ^{6}Li thermal-neutron cross section is about a factor of 6 lower than ^{3}He.

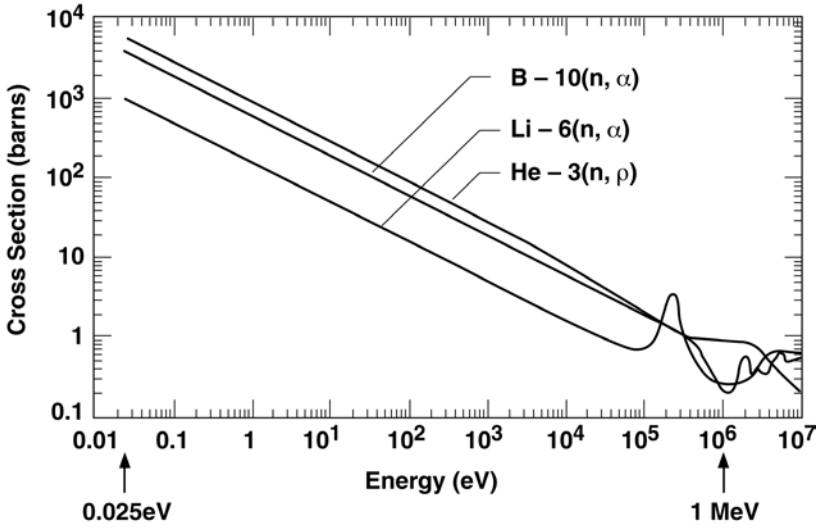

Figure 1: Neutron cross sections as a function of energy for the leading capture reactions being considered for ^{3}He replacement.

The neutron sources of interest for safeguards are from spontaneous fission, induced fission, and alpha,n reactions. The average energy for the source neutrons is 1-2 MeV; however, the high cross-sections for the detector target isotopes are for thermal-energy neutrons. Thus, the detector configuration needs to have a substantial amount of neutron moderator in the immediate vicinity of the detector. Hydrogen is the most efficient isotope to reduce the source energy by scattering, and high density polyethylene (HDPE) is usually the most cost effective method of introducing the hydrogen into the detector moderator area. The first neutron well counter introduced to the IAEA by a member state consisted on a ring of ^{3}He tubes in a plastic bag. The inspectors could carry the lightweight bag system into the plant and add water for the moderator at the point of application. Needless to say, the system was never implemented for inspection.

Figure 2 shows a curve of efficiency versus HDPE thickness for the case where a 1" diameter ^{3}He tube is located in the center of the slab geometry. The efficiency peak is ~ 2.8 cm deep into the poly slab for the tube centerline. Typical thermal-neutron slab detectors have a thickness of 10-13 cm of HDPE or equivalent hydrogen thickness. The hydrogen might be part of the sample matrix, the detector body, or the detector back shield. The hydrogen needs to be in the immediate vicinity of the detection isotope to obtain the full benefit of the hydrogen scattering.

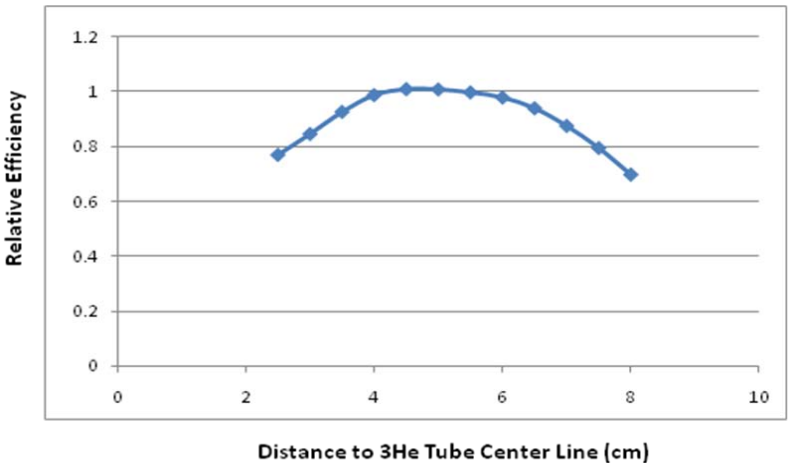

Figure 2: Relative efficiency from MCNPX of a ^{3}He tube (1 inch diameter and 4 atm pressure) embedded in a 11-cm-thick slab of HDPE as a function of depth into the HDPE for fission energy spectrum neutrons.

For a high efficiency neutron slab detector, roughly 1/3 of the incident neutrons diffuse (leak) from the exterior surfaces of the system, ~ 1/3 get captured by the hydrogen in the moderator and Cd liners, and ~ 1/3 provide the useful neutron signal, The leakage loss can be reduced by putting in thicker moderator with detector tubes but the cost and weight increase faster than the efficiency. The parasitic loss to hydrogen capture can be reduced by increasing the density of the ^{3}He (etc.) relative to H; however, the cost increases rapidly with this approach. Because ^{3}He is a nonreactive gas, the ^{3}He has required gas tubes (or equivalent) for containment, and as the tube diameters get smaller and the tube density higher, the cost escalates. This is the primary reason that the fielded ^{3}He tubes are in the 1-2" diameter range.

The competition with H moderator neutron capture is the primary domain where alternatives to ^{3}He gas can make inroads for increased efficiency. The ^{10}B, ^{6}Li, and Gd can exist in the solid and liquid forms where the atom density is much higher than for a gas. As with ^{3}He, the neutron loss from leakage will remove ~ 1/3 of the efficiency for practical sized systems, but the atom density for these replacement isotopes can be much higher than for ^{3}He, thus compensating for the lower cross sections. However, for the solid material forms of Li and boron that are in surface layers, there is the generic problem of getting the reaction products such as alpha particles and ^{7}Li out of the solid medium to provide a signal for the neutron capture. In addition to this problem of charged particle escape probability, there is the lesser problem of thermal-neutron self-shielding in the solid deposit.

To minimize the parasitic loss to hydrogen, the neutron capture isotope such as ^{10}B needs to be intermixed with the hydrogen so that when a thermal neutron is created by H scattering, the ^{10}B captures the neutron before the H can capture it because of the much higher cross section of the ^{10}B, etc. for thermal neutrons.

What About BF_3 Tubes – Neutron detectors using BF_3 tubes have been commercially available for more than four decades. The pluses for the BF_3 detectors are that they are stable, reliable, and more gamma resistant than ^{3}He tubes. On the minus side, they have about half the efficiency of a ^{3}He tube of the same size, and they require a higher bias voltage. The key problem is that the BF_3 gas is hazardous and the current safety requirements will not let them be used in most nuclear facilities. There are research programs underway to develop chemical methods to neutralize any gas leaks by surrounding the tubes

with the chemical absorbent agents. However, the success of this activity is questionable because of the detector cost increase, and the ever escalating accident scenarios.

4. Gamma-Ray Sensitivity

Neutron detectors that are used for safeguards applications need to be resistant to gamma-ray activity, because all nuclear materials emit gamma-rays in excess of the neutron emission. Bulk plutonium samples have associated gamma doses in the range of 0.1-10 R/h (1-100 mSv/h), and the spent fuel gamma dose is ~ 5 orders of magnitude higher. Lead shielding can be used to preferentially shield gammas versus neutrons, but the size and cost increases and the efficiency decreases.

Many of the detectors proposed to be alternatives to ^{3}He, have gamma rejection specifications that are reasonably good; however, the specifications are typically for a small volume detector, and when the size is increased for better efficiency, the gamma rejection gets worse rapidly. Note that the neutron detection efficiency increases slower than the increase in the active detection volume, whereas, the gamma sensitivity increases faster than the square of the volume increase.

For scintillation based detector systems, there is a problem in addition to this gamma pileup effect. The electronics to separate neutron events from gamma events make use of the pulse shape differences between neutrons and gamma rays, so as the volume increases, the pulse rate increase and the system suffers dead time losses. Also, the pulse shape differences between neutrons and gammas tend to blur as the active volume gets larger.

The ^{10}B lined proportional counters, that are the initial focus of the LANL safeguards detectors test program, advertise better gamma-ray rejection capability than for ^{3}He tubes. This is possible because the energy released in the neutron capture reaction with ^{10}B is 1.48 meV for the alpha particle reaction and 0.84 meV for the ^{7}Li ion compared with 0.76 meV for the ^{3}He neutron capture reaction. Also, the ^{10}B proportional detectors are designed with smaller diameter tubes and less gas volume for the gamma-ray induced electron ionization process.

Figure 3 illustrates the Pulse-shape distributions for a ^{3}He tube and for a ^{10}B tube. The primary neutron capture peak at 0.76 meV for ^{3}He is well removed from the electrical noise and the gamma pileup. However, for the ^{10}B solid layer, the desired neutron capture events merge with the noise and gamma pileup. Additives such as Ar in the ^{3}He gas help to bunch the ionization charge collection, and allow the use of short shaping times in the preamplifiers. Figure 4 shows data taken with 254 mm long ^{3}He tube (4 atm) connected to a PDT-10A preamplifier. We see that ^{137}Cs gamma doses up to ~ 100 R/h (1 Sv/h) can be tolerated depending on the neutron source strength. Longer ^{3}He tubes cannot tolerate such a high dose.

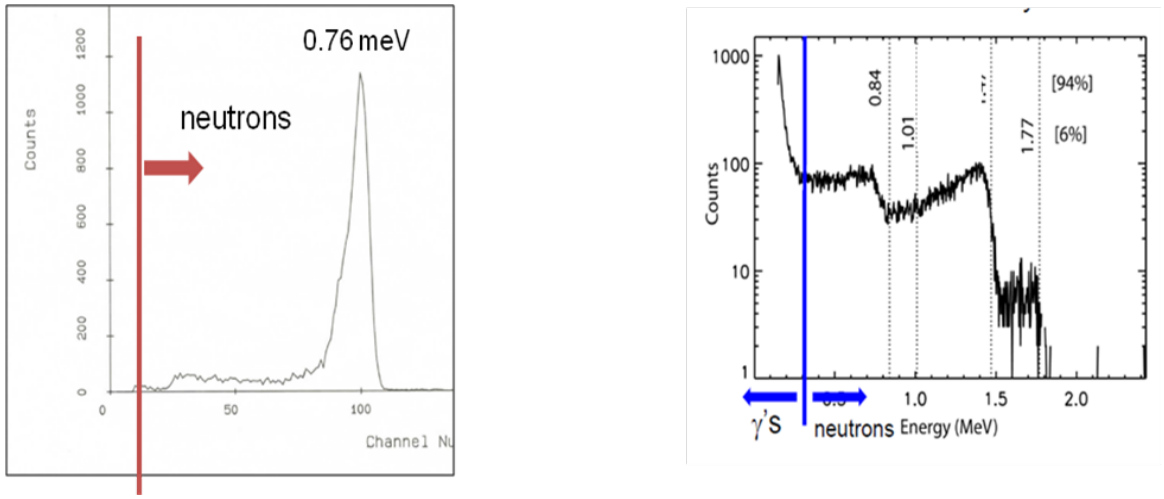

Figure 3: Pulse shape distributions for a ^{3}He tube with 2 μs shaping times (left side) and a thin ^{10}B deposit in an Ar plus methane filled gas tube (right side).

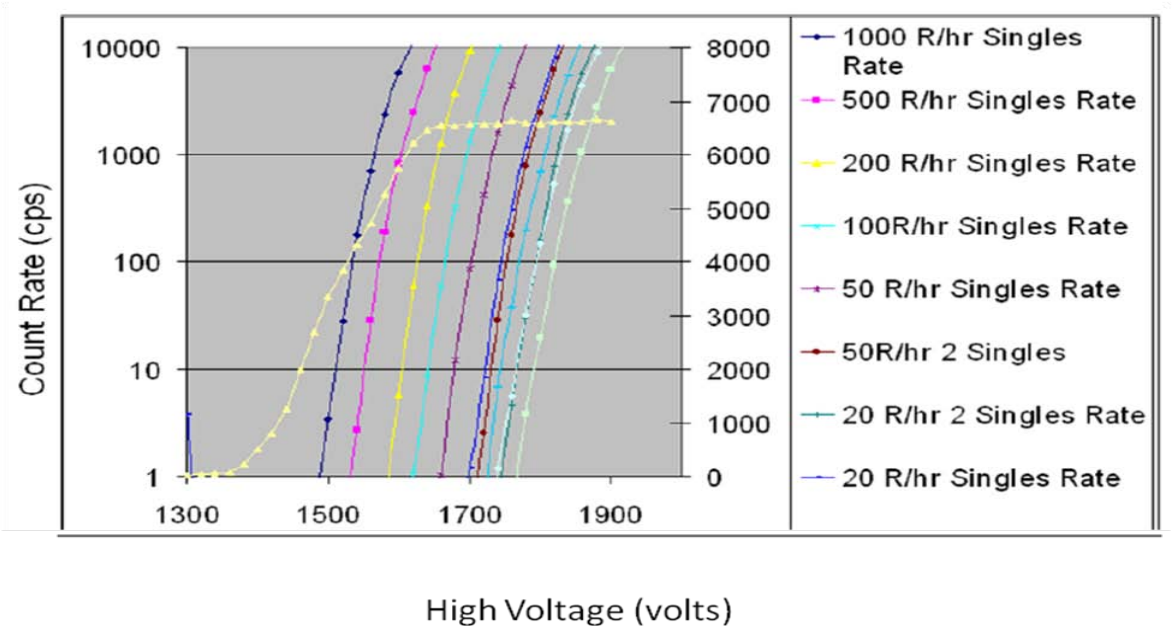

Figure 4: Gamma-ray pileup measurements as a function of high voltage bias for a 25-mm-diameter, 254-mm-long ^{3}He tube at 4 atm pressure.

The LANL test program will expose each of the detectors to gamma doses from ^{137}Cs and/or radium. The gamma pileup problem is a function of both the dose and the detector volume that is exposed to the dose. The gamma dose intensity and area on the face of the detector will be adjusted to provide comparable dose levels for the different detectors as well as the LANL ^{3}He benchmark detector. The gamma/neutron sensitivity ratios will be compared.

5. Die-Away Time

The neutron die-away time is the time for a neutron to slow down from fission spectrum energy to thermal-neutron energy and to be captured by the ^{3}He or hydrogen or to diffuse from the system. In most thermal detectors the neutron population decreases nearly exponentially in time. The time constant is called the die-away time.

The time to go from the fission energy to a few eV requires only ~ 1-3 us; however, the scattering time spent at near thermal-neutron energies is much longer (~ 5-100 us). The effective die-away time of the detector is more a function of the neutron moderating material and geometry than of the capture reaction isotope. For typical high counting rate applications in safeguards, the doubles measurement precision improves with the sqrt of the die-away time. Thus, a factor of 4 decrease in the die-away time provides a factor of two improvement in the statistical precision.

A figure of merit in evaluating neutron detector options for potential replacement of ^{3}He detectors is the efficiency divided by the sqrt of the die-away time. This ratio is directly proportional to the statistical precision for the doubles counting rate.

6. Detector Dead Time

Relatively high efficiency is required for neutron counters used in safeguards applications to accommodate neutron coincidence counting. However, the neutron emission rate from bulk plutonium samples is high. A 2 kg sample of PuO_2 emits several million neutrons per second, so a 30% efficient detector would have a counting rate in excess of 1 MHz. The neutron yield from spent fuel and impure samples can be an order of magnitude higher because of the ^{244}Cm and (alpha,n) reactions. The dead-time in ^{3}He tubes is relatively large because of the charge collection time in the gas tube. The larger diameter tubes are slower than the smaller diameters. Additives such as Ar and CF_4 are added to the ^{3}He gas to help reduce the ionization charge collection time.

A reduction in dead-time is one area where many of the alternative technology to ^{3}He gas tubes can outperform the ^{3}He gas tubes. The ^{10}B lined detectors can be more than five times faster than ^{3}He tubes because the cathode (walls) to anode (wire) spacing is significantly reduced to allow for more surface area for the ^{10}B layer, and the shift from ^{3}He gas to Ar to provide the ionization of the neutron reaction products.

The light scintillation detectors with the associated photomultiplier tubes are more than an order of magnitude faster, and have less dead-time. However, many of the methods to separate neutron and gamma-ray events make use of pulse-time analysis that introduces a new source of dead-time.

7. Survivability

Neutron detectors that are used in safeguards applications must be able to function for long periods (years) under the continuous irradiation of both neutrons and gamma-rays without a degradation in performance. The potential source of the radiation is the nuclear material itself. The bulk plutonium oxide samples emit on the order of 1e+6 n/s and a higher number of gamma rays. The surface gamma dose for reactor grade plutonium is in the range of 0.1-1 R/h and much higher for spent fuel materials. Lead shielding can be used for some applications, but then the cost, weight, and safety issues increase.

The use of ^{3}He gas tubes has been relatively immune of this problem in the past because of the inert nature of the noble gas. Potential replacement detectors that make use of optical scintillation light collection will have to deal with the light transmission properties of the measured signal. Gas proportional counters such as ^{10}B and ^{6}Li lined gas ionization counters will be more robust related to radiation damage problems.

In all cases, the support electronics that are collocated with the neutron detector must survive a similar radiation exposure. The ^{3}He gas tubes have used the AMPTEK A-111 amplifier or the PDT-10A amplifiers for more than two decades with gamma dose survivability for integrated dose levels of more than 10-100 Mega Rad. However, the original versions of these amplifier circuits were modified to replace radiation sensitive components after failing the original survivability tests.

8. Scalability Considerations

Before alternative neutron detectors can make a dent in the demand on ^{3}He gas supply, the detectors need to be scalable to the larger geometries that use the lion's share of the ^{3}He gas. Small neutron tubes such as spent fuel discharge monitors use less than a liter of gas, and their replacement does not address the problem. The larger, more efficient, safeguards detectors such as slab and well detectors require ^{3}He gas in the range of 50 to 500 liters per unit.

There are several challenges that need to be overcome relating to scalability including:

1) Total package size to get the required efficiency
2) Signal collection attenuation versus size (primarily a scintillation system problem)
3) Gamma sensitivity that increases with volume much faster than the neutron efficiency
4) Cross-talk between multiple subcomponents that are introduced for higher efficiency
5) Complex electronics that require operation by a specialist
6) Robustness and reliability versus system complexity
7) Commercial production cost for a large scale system

The neutrons are born as fast neutrons with a fission energy distribution (1-2 MeV). However, the neutron detection reactions require thermal neutrons to take advantage of the high thermal-neutron reaction cross sections. The leading thermal-neutron reaction targets are shown in Fig. 1 where ^{3}He has the highest cross section, but ^{10}B and ^{6}Li are possible replacement candidates.

To be scalable, the neutron detectors need to have a high ratio of neutron detection active volume to the total system volume. Without this feature, the total detector size gets to be too large to fit into the available space for in-plant and portable applications. Figure 5 shows a ^{6}Li based scintillation multiplicity detector [2] and a High Level Neutron Coincidence Counter (HLNC) [3]. Both of these systems were developed at LANL and measure ^{240}Pu effective mass via passive neutron multiplicity counting. The efficiency of the HLNC is higher than the scintillation based system; however, the die-away time of the

scintillation detector is about six times shorter. Thus, the multiplicity statistical error for the scintillation system is about a factor of two better than for the HLNC for samples with high (alpha,n) rates. However, the larger system was never implemented because of the size, cost, and complexity.

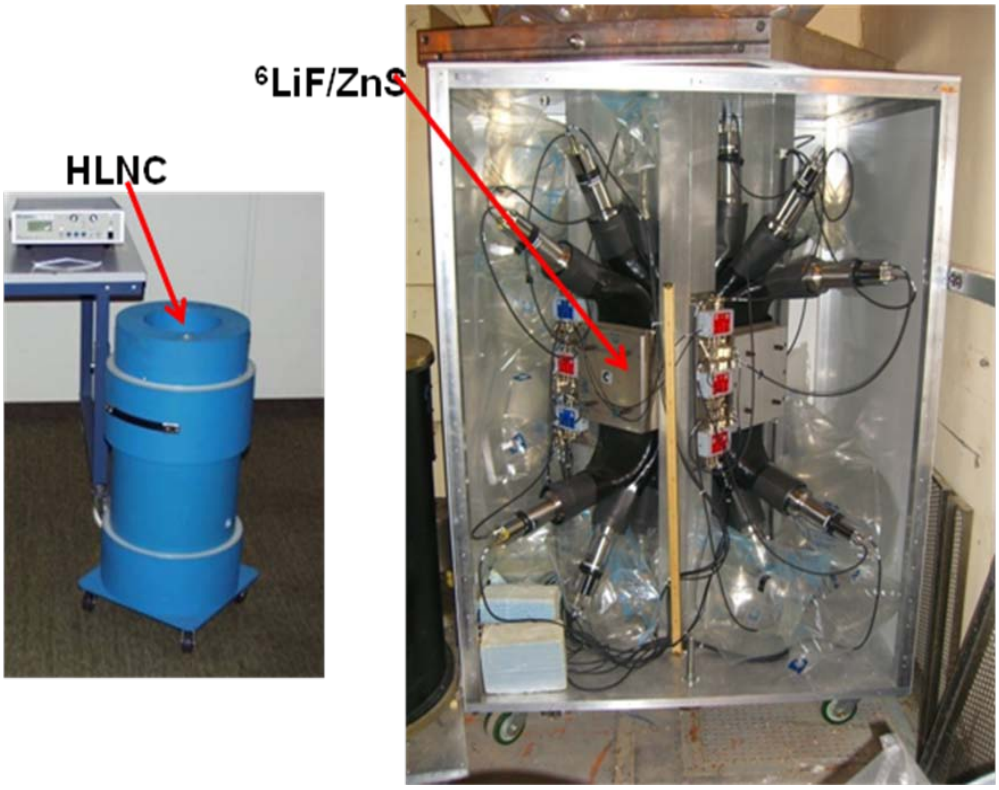

Figure 5: The compact ^{3}He based HLNC Detector (left side), and ^{6}LiF/ZnS scintillator based system (right side) to illustrate the scalability problem.

9. Conclusions

The detector test program at LANL will focus on those parameters that are of key importance for the future application of ^{3}He replacement detectors for safeguards applications. The efficiency and stability of the potential replacement detectors will be evaluated. Also, the less obvious parameters such as die-away time, dead time, and gamma sensitivity will be measured and compared with ^{3}He. The robustness of the detectors to radio frequency and micro phonic noise interference will be tested, and all of the parameters will be compared with ^{3}He tube based systems.

To make a significant reduction in the demand on ^{3}He gas, the replacement detectors need to be scalable to the large sized ^{3}He systems that are currently used for slab detectors and well detectors. The nuclear facilities have limited space for safeguards related NDA systems, and the floor space is very costly. Thus, a high efficiency density is needed for the replacement systems, and of equal importance are reliability and robustness, and there are stringent seismic safety regulations. The future of neutron detection for safeguards applications is moving towards installed equipment operating at a 100% duty cycle, and

reliability and survivability are essential. The ^{3}He tubes are the only detector option that can currently meet these requirements. However, potential replacement technologies are under development. The safeguards community recognizes the shortage issue and is working towards a solution.

There is a viable commercial market for neutron detectors that are alternatives to ^{3}He, and vendor participation in the development is an indication of the potential promise in the proposed technology.

10. Acknowledgements

This work was funded by the Next Generation Safeguards Initiative (NGSI), Office of Nonproliferation and International Security (NIS), National Nuclear Security Administration (NNSA).

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The Passive Neutron Enrichment Meter for Uranium Cylinder Assay

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Abstract:

As fuel cycle technology becomes more prevalent around the world, international safeguards have become increasingly important in verifying that nuclear materials have not been diverted. Uranium enrichment technology is a critical pathway to nuclear weapons development, making safeguards of enrichment facilities especially important. Independently-verifiable material accountancy is a fundamental measure in detecting diversion of nuclear materials. This paper is about a new instrument for uranium cylinder assay for enrichment plant safeguards called the Passive Neutron Enrichment Meter (PNEM). The measurement objective is to simultaneously verify uranium mass and enrichment in UF_6 cylinders. It can be used with feed, product, and tails cylinders. Here, we consider the enrichment range up to 5% ^{235}U. The concept is to use the doubles-to-singles count rate to give a measure of the ^{235}U enrichment and the singles count rate to provide a measure of the total uranium mass. The cadmium ratio is an additional signature for the enrichment that is especially useful for feed and tails cylinders. PNEM is a ^{3}He-based system that consists of two portable detector pods. Uranium enrichment in UF_6 cylinders is typically determined using a gamma-ray-based method that only samples a tiny volume of the cylinder's content and requires knowledge of the cylinder wall thickness. The PNEM approach has several advantages over gamma-ray-based methods including a deeper penetration depth into the cylinder, meaning it can be used with heterogeneous isotopic mixtures of UF_6.

In this paper, we describe a Monte Carlo modelling study where we have examined the sensitivity of the system to systematic uncertainties such as the distribution of UF_6 within the cylinder. We also compare characterization measurements of the PNEM prototype to the expected measurements calculated with Monte Carlo simulations.

Keywords: UF_6; cylinder; enrichment; PNEM

1. Introduction

As fuel cycle technology becomes more prevalent around the world, international safeguards have become increasingly important in verifying that nuclear materials have not been diverted. Uranium enrichment technology is a critical pathway to nuclear weapons development, making safeguards of enrichment facilities especially important. Independently-verifiable material accountancy is a fundamental measure in detecting diversion of nuclear materials. This paper is about a new instrument for uranium cylinder assay for enrichment plant safeguards called the Passive Neutron Enrichment Meter (PNEM). The objective is to simultaneously verify uranium mass and enrichment in UF_6 cylinders. It can be used with feed, product, and tails cylinders. Here, we consider the enrichment range up to 5% ^{235}U.

Because the in-process inventory is very small, the majority of the UF_6 at an enrichment plant is contained in 30B and 48Y cylinders. 30B cylinders have a 30-in. diameter, 1/2-in.-thick steel wall, and hold up to 5% enriched UF_6 (i.e., product). 48Y cylinders have a 48-in. diameter, 5/8-in.-thick steel wall, and hold natural and depleted UF_6 (i.e., feed and tails).[1]

Traditionally, the International Atomic Energy Agency (IAEA) has used a portable load-cell-based system (LCBS) to verify uranium mass in UF_6 cylinders and a gamma-ray technique that measures the net counts in the 186-keV peak from ^{235}U to verify enrichment.[2,3] In combination, these two methods give a measure of the ^{235}U content of a cylinder; however, there are several drawbacks to both. The LCBS is time consuming to use, requires a valid tare weight for each cylinder, and there is no indication of whether the material inside the cylinder is, in fact, nuclear material. The 186-keV gamma ray can only penetrate a small distance in UF_6, making it difficult to get a representative sample of a heterogeneous isotopic mixture. The gamma-ray technique also requires a measurement of the cylinder wall thickness to correct for attenuation in the steel.

The objective of safeguards is the timely detection of the diversion of a significant quantity of special nuclear material. Current safeguards methods for uranium enrichment were designed for plants with significantly smaller throughput than the modern commercial plants.[4] In order to verify that the safeguards objectives are met in these larger plants, the IAEA is calling for increasingly more accurate assay techniques that can be performed near real time. Strained human resources are also pushing safeguards technologies towards more unattended monitoring systems in order to make better use of inspector time. The PNEM technique has the potential to be used in lieu of the load cell/gamma-ray combination or, alternatively, as a cross check of those and other monitoring systems in the plant. Neutron-based cylinder assay is highly complementary to the traditional load cell and gamma-ray spectroscopy methods. For example, neutrons provide deep penetration into the cylinder, meaning they are well suited for assaying heterogeneous isotopic mixtures (common in tails cylinders where the 186-keV peak from ^{235}U is weak). Passive neutron methods are also readily adaptable to unattended mode operation, where they can be used as part of an attribute monitoring system to check for consistency among multiple types of sensors. Furthermore, there may be other applications where the portability of the PNEM detector pods is an asset during on-site inspections.

The primary neutron sources in UF_6 are (α,n) neutrons from ^{234}U alpha bombardment of fluorine and ^{238}U spontaneous fission. In general, the enrichment of ^{234}U follows that of ^{235}U, so the random (α,n) source can be related to ^{235}U. Recently, a system called the Uranium Cylinder Assay System (UCAS) was installed at Rokkasho Enrichment Plant in Japan that uses total neutron counting to determine the uranium mass in 30B and 48Y cylinders.[5] UCAS was designed to be an operator system, as opposed to an inspector system, and relies on *a priori* knowledge of the enrichment and $^{234}U/^{235}U$ ratio. The PNEM system builds on the UCAS approach by adding coincidence counting to independently verify the ^{235}U enrichment. It makes use of induced fission in ^{235}U from the thermal neutron return from the detector. The concept is to use the doubles-to-singles ratio to give a measure of the ^{235}U enrichment and the singles count rate to provide a measure of the total uranium mass. The cadmium ratio is an additional signature for the enrichment that is especially useful for feed and tails cylinders.

In the following sections, we describe the design of PNEM and how the prototype system will interface with a UF_6 cylinder. We also describe a Monte Carlo modelling study where we have examined the sensitivity of the system to systematic uncertainties such as the distribution of UF_6 within the cylinder. All of the physics calculations were performed using the transport code Monte Carlo N-Particle Extended (MCNPX). Finally, we compare characterization measurements of the PNEM prototype to the expected measurements calculated with MCNPX simulations.

2. Mechanical and Electrical Design

PNEM is a ^{3}He-based system. The prototype was designed to be a portable instrument with two briefcase-sized detector pods. Both pods weigh approximately 20 kg and have a handle on one end for carrying. They each have two rows of six ^{3}He tubes, where the tubes have a 2.54 cm (1 in.) diameter, 50.8 cm (20 in.) active length, and 4 atm of ^{3}He pressure. The position of the tubes was optimized using a figure of merit to minimize the statistical uncertainty in the detector.[6] A photo of one of the detector pods is shown in Figure 1.

In order to minimize the length and weight of the pods, Precision Data Technology (PDT) designed a compact electronics package for each PNEM pod, shown in Figure 2. The amplifier is lower profile than the standard amplifiers typically used with ^{3}He tubes. There are three output signals for each system: one for the front row of tubes, one for the back row of tubes, and one sum total output from all twelve tubes.

Figure 1: PNEM detector pod.

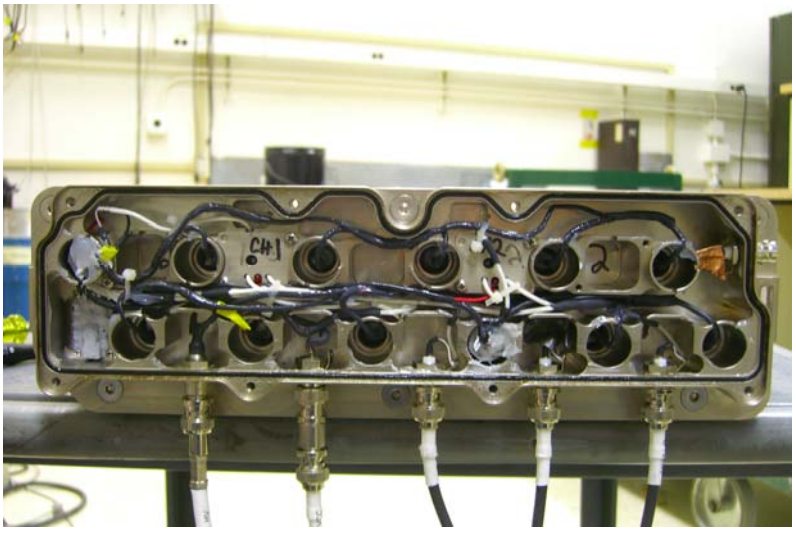


Figure 2: PNEM electronics package.

The conceptual measurement position of the prototype detector pods for a 30B cylinder is shown in Figure 3 (cylinder cradle not shown). The measurements can be done in the storage area of an enrichment plant by placing the detector pods on the floor on either side of the cylinder. We assumed that the cylinders sit on a cradle approximately 10 cm above the floor. The detectors should be positioned near the bottom of the cylinder because that is where the bulk of the UF_6 is most likely to be located. The technique does not rely on the particular pod-cylinder orientation shown in Figure 3 or the form factor of two small slab detectors. An installed PNEM system could be made for an enrichment plant where the pod positioning was, for instance, directly underneath the cylinder. The data analysis technique (i.e., doubles-to-singles ratio or cadmium ratio for enrichment and total neutron counting for uranium mass) could be used to assay smaller 1S UF_6 cylinders using a well counter. This concept has been explored at Los Alamos National Laboratory using a high efficiency, four-ring well counter.[7]

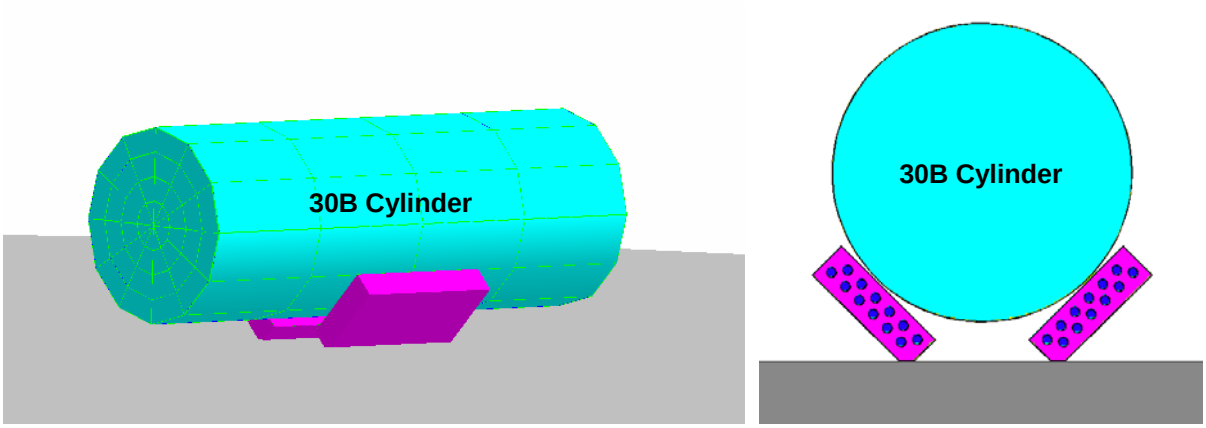


Figure 3: PNEM measurement position with respect to a 30B cylinder.

3. Physics Calculations

All of the physics calculations were performed using the transport code MCNPX. The (α,n) neutron energy spectrum and (α,n) and spontaneous fission source strengths were calculated using another code called SOURCES 4C. As shown previously by Miller et al., a full 30B cylinder containing low-enriched UF_6 will achieve 1% counting statistics in fifteen minutes with the prototype 4 atm system.[6] If the ^3He tubes were replaced by tubes with 10 atm of gas pressure, the singles efficiency would increase by 16% and doubles by almost 40%.

The biggest source of systematic uncertainty for PNEM is the distribution of UF_6 within the cylinder. The geometry effects are more pronounced in 30B cylinders where multiplication plays a bigger role in the doubles count rate than in 48Y cylinders. The UF_6 profile inside the cylinder depends on how the cylinder was filled and the storage conditions. Berndt, Franke, and Mortreau used the filling profiles shown in Figure 4 in their modelling study of geometry effects on a theoretical total neutron counter for UF_6 cylinders.[8] The x-factor describes the percentage of UF_6 covering the inner cylinder wall with a layer of constant thickness.

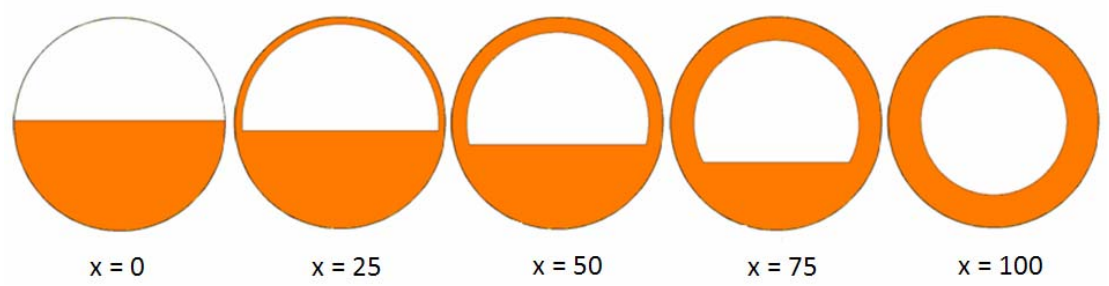
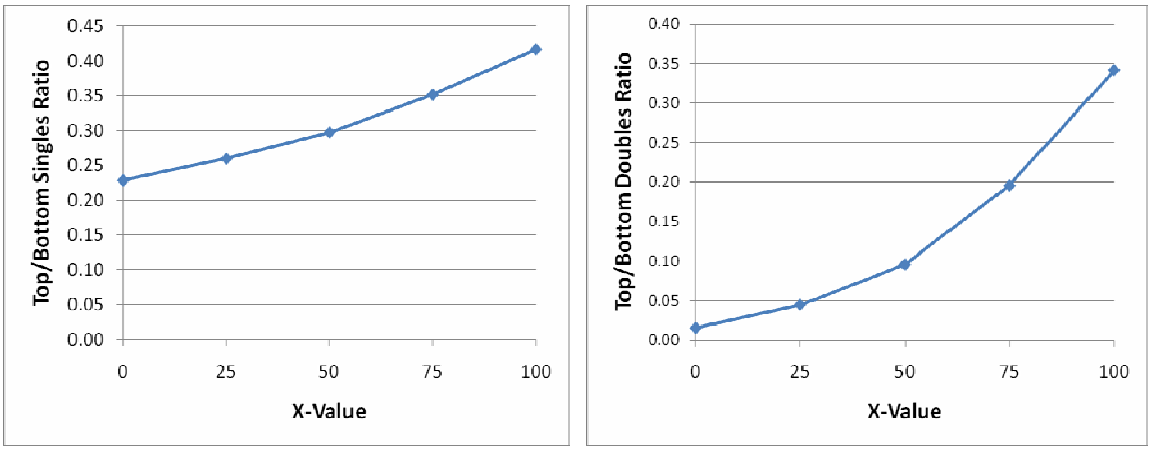


Figure 4: UF_6 filling profiles.

Feed cylinders containing natural UF_6 from conversion plants are filled in liquid phase, meaning the UF_6 collects at the bottom of the cylinder. This is illustrated by the x=0 case. Product and tails cylinders are generally filled by desublimation, where solid UF_6 adheres evenly to the cylinder wall, creating an annular ring. This is illustrated by the x=100 case. Over time, the UF_6 on the upper part of the wall will slough off and fall to the bottom (x=25, 50, and 75).

The filling profiles shown in Figure 4 represent the extreme bounding cases. In practice, the true range of filling profiles for most cylinders is a smaller subset of Figure 4. The size of that subset is something that needs further study. To get a better understanding of the true range of filling profiles, measurements should be taken on a large population of cylinders. This type of measurement campaign would help quantify the systematic uncertainty associated with the distribution of UF_6 inside 30B and 48Y cylinders.

Preliminary MCNPX modelling results show that a signature for the x-factor, or filling profile, can be obtained by placing a third detector pod on top of the cylinder and looking at the ratio of the top-to-bottom pods. We modelled a 30B cylinder with the three-pod configuration with each of the five filling profiles shown in Figure 4. A plot of the results is shown in Figure 5. Field trials of the PNEM system may benefit from this additional detector pod to better understand the variability in source distribution between cylinders, but the third pod would not be part of a deployed PNEM system.

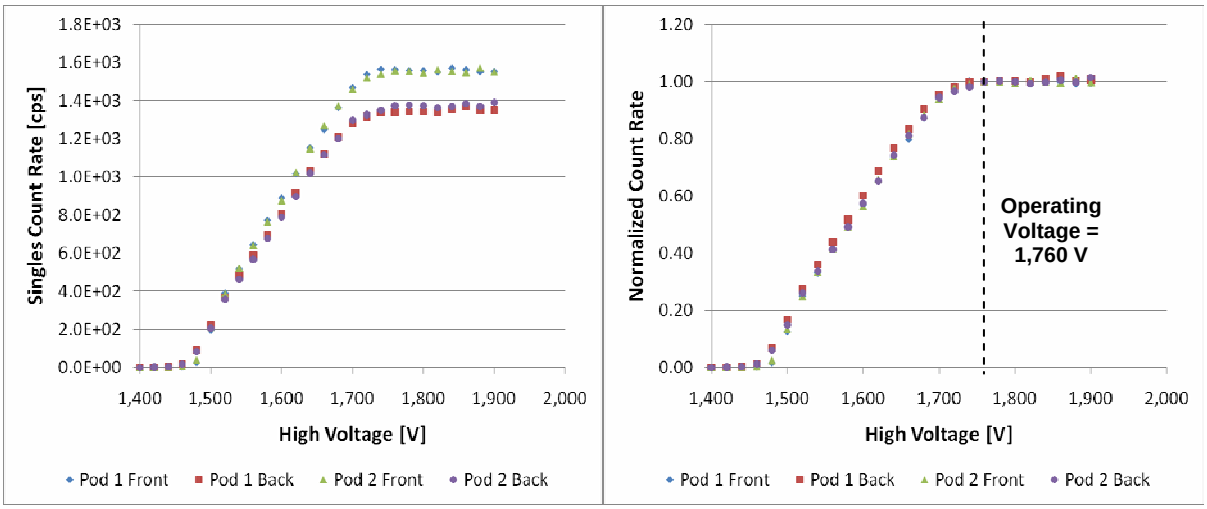


(a) (b)

Figure 5: Ratio of top-to-bottom pods as a function of the x-factor for (a) singles and (b) doubles count rates.

4. Characterization Measurements

Before field trials are done, it is customary to perform characterization measurements on a new instrument to ensure that the detector is working as expected and to benchmark computational models. We started the characterization measurements by performing a number of tests on the electronics of the PNEM detector pods. This included creating voltage plateaus and gain matching the ^{3}He tubes. To collect the voltage plateaus, we placed each pod on a metal cart with a ^{252}Cf source centred on the front face of the pod. Using INCC, the standard coincidence counting software, we took counts on Pod 1 in 20 V increments between 1,400 and 1,900 V. The results are shown in Figure 6. Figure 6(a) shows that the count rate in the front tubes is higher than the back tubes, as expected. To establish an operating voltage that provides maximum stability, we chose the operating voltage at 1,760 V, which is 40 V above the “knee” of the curve in Figure 6(b), which shows the normalized count rates. The gain, or voltage amplitude for a given event in the detector, of Pod 2 was matched to Pod 1. The electronics were also checked for stability, noise, and sensitivity to moisture.



(a) (b)

Figure 6: PNEM voltage plateaus showing (a) absolute measurements and (b) normalized count rates.

The remaining measurements were performed to characterize the detector itself. The first of these was a series of identical measurements with shift register gate widths of 16, 32, 64, and 128 μs to determine the die-away time in the detector. The die-away time (τ) is the average neutron lifetime in a detector and is determined primarily by the size, shape, composition, and efficiency of the counter. It can be calculated for doubles count rates D_1 and D_2 and gate widths G_1 and G_2, where G_2 is twice G_1, using the following equation:

$$\tau = -\frac{G_1}{ln(D_2/D_1 - 1)}$$

The average die-away time in the PNEM system is 44 μs. The gate width for the remaining measurements was chosen to minimize the uncertainty in the doubles count rate. Using the die-away time measurements, we plotted the percent uncertainty in the count rate as a function of gate width. Figure 7 shows that the optimal gate width for PNEM is 64 μs. Although deadtime should not be a major factor in UF_6 cylinder measurements, we also calculated the deadtime parameters using the twin source method.

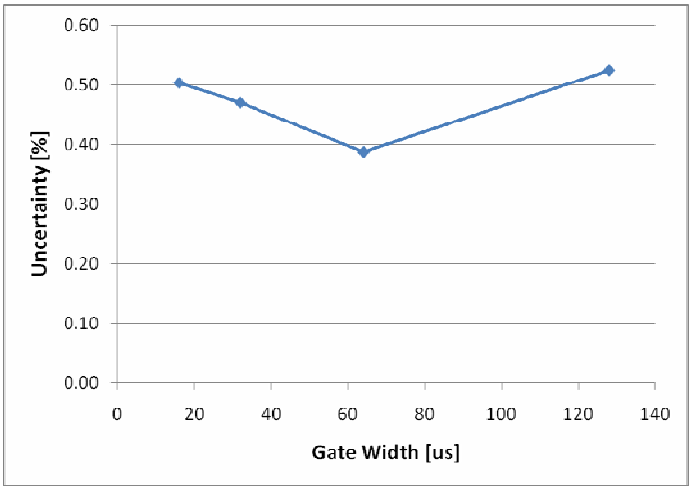


Figure 7: Percent uncertainty in the doubles count rate as a function of gate width.

The last series of measurements was performed to characterize the detector response profiles and benchmark the MCNPX calculations. Again, we used a ^{252}Cf source. With the source centred 30 cm away from the front face, we found the efficiency of a single pod is 1.9%. Using the same single-pod setup, we created response profiles in the x-, y-, and z-directions by taking measurements of the source at 5 cm increments along each axis. The measured profiles were used to benchmark MCNPX simulations of the same setup. The pods were then put into the proposed 30B measurement configuration shown below.

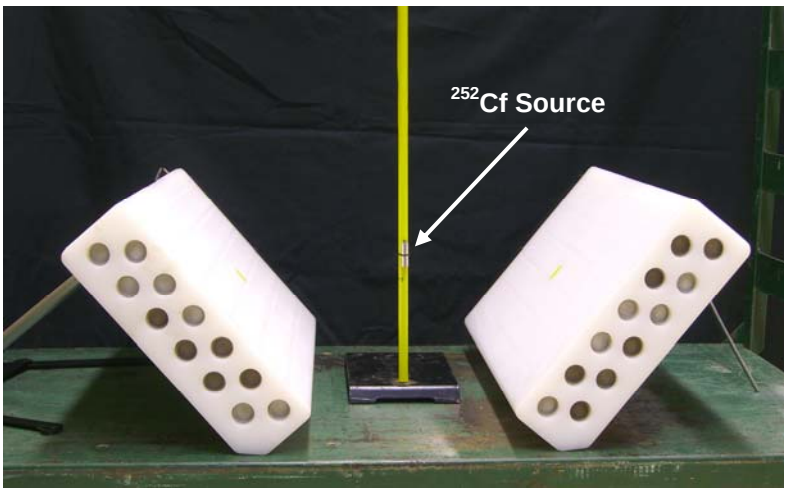
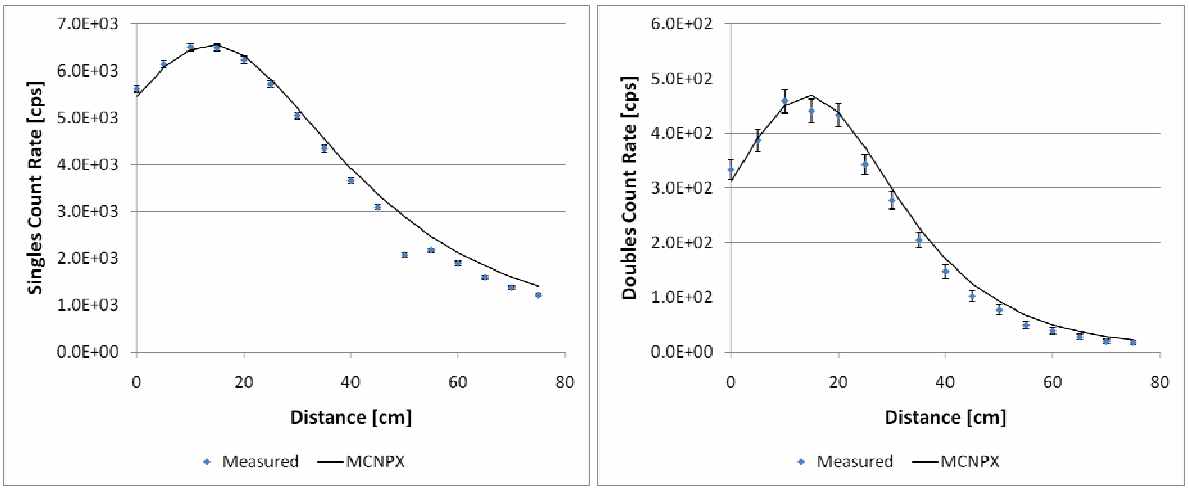


Figure 8: Photograph of the PNEM detector pods in the proposed 30B measurement position.

We created a vertical response profile for the two-pod configuration by taking measurements from 0 to 80 cm above the floor in 5 cm increments. Figure 8 shows the ring stand and ^{252}Cf source that was used for this measurement. The ring stand was centred between the two pods and along the length of the ^{3}He tubes. The measurements and MCNPX modelling results are shown in Figure 9 for the singles and doubles count rates. The distance is given in centimetres above the floor. Both sets of data show good agreement between the measurements and simulations, especially in the region closest to the floor. The MCNPX results predicted slightly higher than measured count rates when the source was above the detector pods. This is likely due to room effects not included in the simulation.



(a) (b)

Figure 9: Measured and calculated response profiles for the (a) singles and (b) doubles count rates.

5. Summary & Future Work

To summarize, we have described a new instrument and data analysis technique for uranium cylinder assay called PNEM. It is a ^{3}He-based passive neutron detection system, and the measurement objective is to simultaneously verify mass and enrichment of UF_6 inside 30B and 48Y cylinders. In this paper, we described the mechanical and electrical design of the prototype PNEM detector pods as well as the proposed measurement position with respect to a 30B cylinder. MCNPX and SOURCES 4C were used for physics calculations. We used the codes to explore a technique to determine the distribution of UF_6 within the cylinder, which is the technique's largest source of systematic uncertainty. Finally, we described ^{252}Cf measurements that were performed at Los Alamos National Laboratory to characterize and test the prototype PNEM system. We found good agreement between the ^{252}Cf measurements and MCNPX simulations of the measurements, which helps lend credibility to the UF_6 cylinder simulations.

The next step in this work will be a field test of UF_6 cylinders in a uranium enrichment plant. We also plan to conduct additional MCNPX simulations to better understand systematic uncertainties associated with parameters such as the ^{234}U content of the UF_6 as a function of enrichment.

6. Acknowledgements

The funding for this work was provided by the Next Generation Safeguards Initiative (NGSI), Office of Nonproliferation and International Security (NIS), National Nuclear Security Administration (NNSA).

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Digital signal processing equipment for neutron coincidence counting

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Abstract:

Determination of detector parameters is essential for precise measurements. The PTR multichannel list mode device has some features for this.

One of the most basic characteristics of a neutron detector is the high voltage plateau. PTR is capable to automatic measure high voltage dependency of count rate. This dependency can be plotted even channel by channel.

By unfolding the follow-up time distribution into channel files the efficiency contribution of each channel can be calculated. Having channel rates even ring ratios can be calculated. The shape of channel files helps to set predelay value.

Rossi-alpha distribution and die away time can be determined for each channel file. Die away time values help to determine optimum gate length.

Multichannel list mode devices can be used to check the functionality of individual channels. Data coming from suspicious channels can be simply subtracted instead of repeating the whole measurement.

Measurements with the widely used one channel HLNC and a multichannel detector built at JRC, Ispra are presented to illustrate the use of PTR devices.

Keywords: neutron coincidence, list mode, follow-up

1. Introduction

Neutron coincidence counting has been well established in recent decades. The method was developed for determining effective Pu-240 content of plutonium containing samples and it has been regularly used in safeguards measurements.

Rapid development of computational hardware made possible to build instruments capable of saving raw data on mass storage devices [1]. This technique is called list mode. These devices describe each pulse by a time value. A wide spread method is to measure the time elapsed since beginning of measurement. This is called time stamp and describes each pulse by a high precision number. Another method is to save only the elapsed time since the previous pulse or follow-up time. This method requires less space as values to be stored are always of limited magnitude. In either case the sequence of values is written to a mass storage device.

Multiplicity counting involves the use of a high number of detector tubes. The outputs of all tubes are added together by a simple logical OR operation. In this way impulses which are too close time will be melted together. The loss is a considerable part of the overall impulse rate in the high range. This and

other error sources are compensated numerically by the dead time correction. To reduce the necessary compensation and to rise the upper limit for overall measuring rate new devices have several independent digital input channels and merge channel data with digital, loss-free processing.

Some of the recent developments save also the channel number along with time data. This enables several new features. The following section presents a device which offers all these features. After that some cases are shown where list mode or channel number handling capability or both are of particular interest.

2. Pulse Train Recorder PTR-32

Pulse Train Recorder is a multichannel list mode device with channel handling capability. It was developed in the Institute of Isotopes. A comparative testing at Los Alamos National Laboratory gave good results [2].

The device accepts standard TTL pulses on 32 channels coming from a neutron detector. Maximal accepted impulse rate is two and a half million cps. Collected data is sent through a USB line to a PC and stored in a binary file. Data files can be evaluated after measurement.

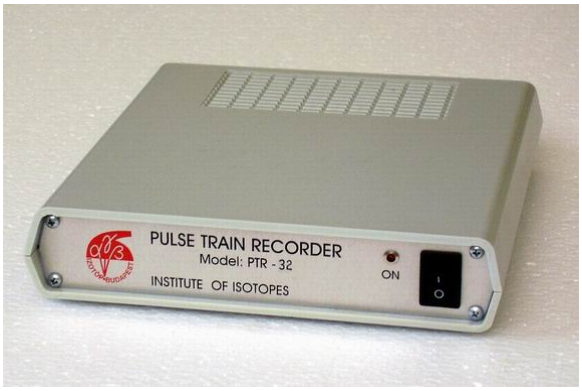

Fig. 1 Pulse Train Recorder device

PTR-32 works with follow-up times instead of time stamps. Time intervals between successive neutron pulses are measured as number of clock periods passed. Impulses arriving on different channels in the same clock period are also accepted. They are saved in increasing order of channel number. The first one gets the follow-up time since the last pulse before and the others are described by a zero as there was no clock pulse between them. Follow-up times are integer numbers. Resulting data files contain the same information as with time stamping but require less space and can be processed faster.

Digitizing causes impulse loss if there are more incoming impulses in a clock period. Derandomizing circuits compensated for this by a fast intermediate storage which delayed excess impulse processing until the next free clock period. High clock rate of PTR-32 make this unnecessary.

Data acquisition is controlled by a program running under Windows. It displays continuously follow-up distribution while data acquisition. The interactive graph is expandable or collapsible and this helps to identify possible measuring problems or interesting features in the data. Data files have a header block containing automatically filled-in and user entered information. The program can read back previously recorded binary data files and graphs.

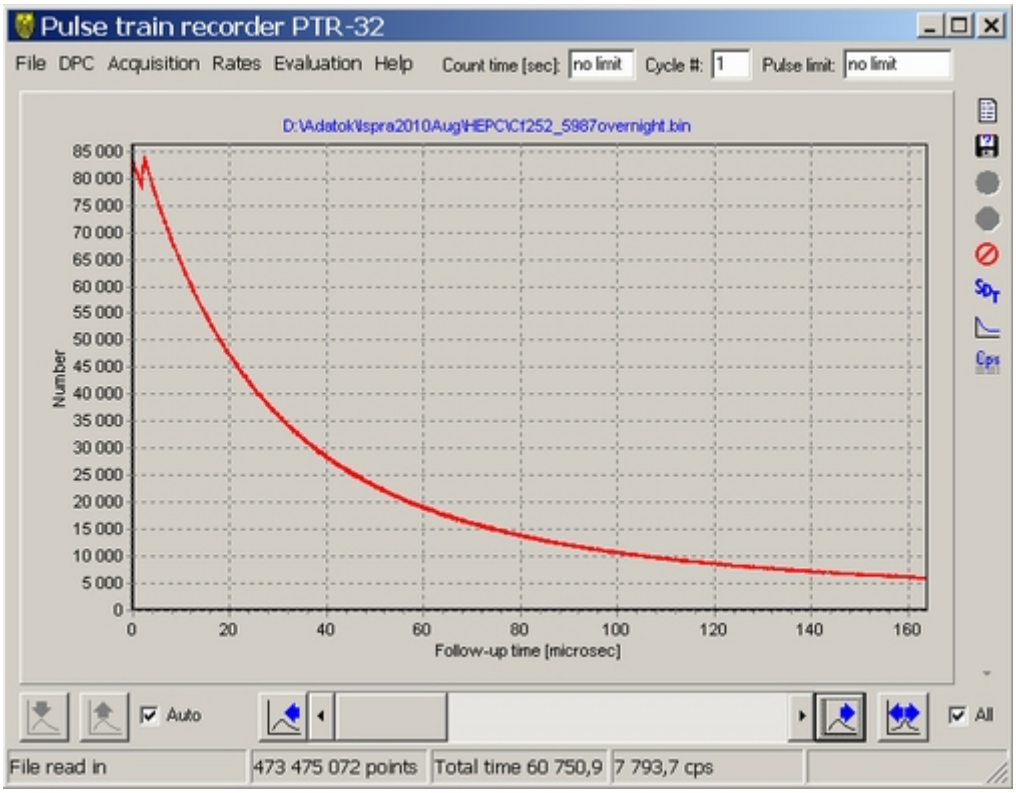

Fig. 2 PTR-32 main window

2.1 Coincidence rate calculation

Coincidence rates are calculated by the formulas of the point model. Calculations are made in a fractional part of the acquisition time. Processing time never exceeds a few percent of acquisition time even at high count rates. The Neutron program took part in the Neutron Coincidence Benchmark Test and outraged with its speed [3].

Predelay, gate width and long delay can be set prior to calculation. The program can handle virtual unlimited high multiplicity numbers. The same data set can be evaluated with different parameters. Distribution values and calculation results can be saved in a text file.

2.2 Calculation of Rossi-α distribution

Rossi-α distribution describes the detection probability of another neutron after a trigger event in function of time. Random events have in this approach a uniform distribution whereas fission neutrons are time correlated that is usually described by a single exponential term. Calculating this distribution consumes much processor time even with integer arithmetic, that is why it is done by a separate program.

Die-away calculation is made by fitting $N(t) = A + R \cdot e^{-t/\tau}$. The calculated distribution is 1024 μs long with 100 ns time bins. Distribution values and calculation results can be saved in a text file.

3. High voltage plateau

Manufacturers specify the recommended high voltage for their detectors. It is usually near to the low end of the high voltage plateau. This should be checked from time to time. PTR-32 offers an automated process for it. A well maintained detector should have a HV plateau like in Fig. 2. The yellow marked excess pulse region will be discussed below.

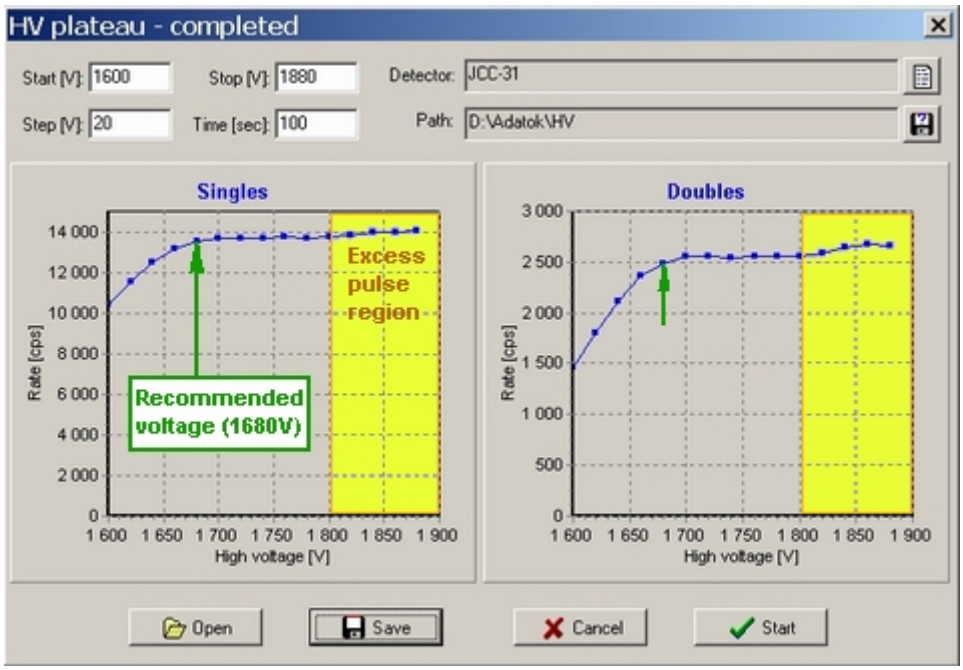

Fig. 3 High voltage plateau

If for some reason the instrument is not properly set, the plateau can be distorted like in Fig. 4.

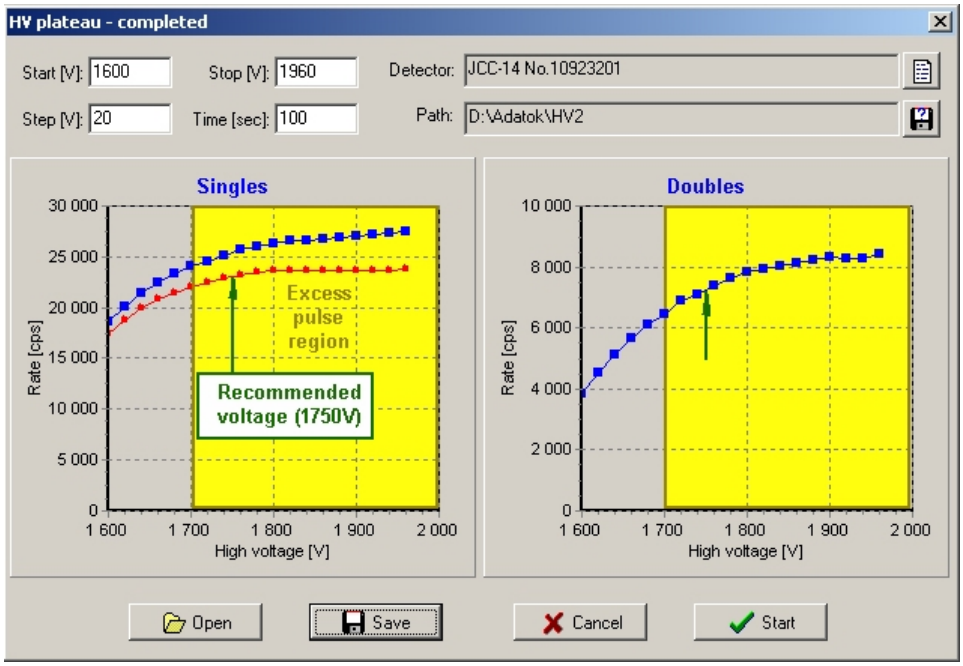

Fig. 4 Distorted plateau

This can be simply explained by the follow-up distribution always shown by the PTR-32 software. Fig. 5. shows several follow-up distributions with high voltage as parameter. The distribution develops false excess pulses produced by the electronics around 1 µs with increasing high voltage. A properly working detector has excess pulses only well above the recommended high voltage, as shown in the figure below. The high voltage region where excess pulses are occurring is marked with yellow background in Figs 3 and 4. Excess pulses give a moderate lift to the overall impulse rate and therefore also the Doubles rate. As Fig. 4 indicates excess pulses appear before the recommended high voltage for that detector and the lifting effect distorts the whole plateau.

For proving the lifting effect of excess pulses impulses with less than 3 µs follow-up time were removed from the impulse train. The curve with small red dots in Fig. 4 clearly shows that this operation restores the plateau.

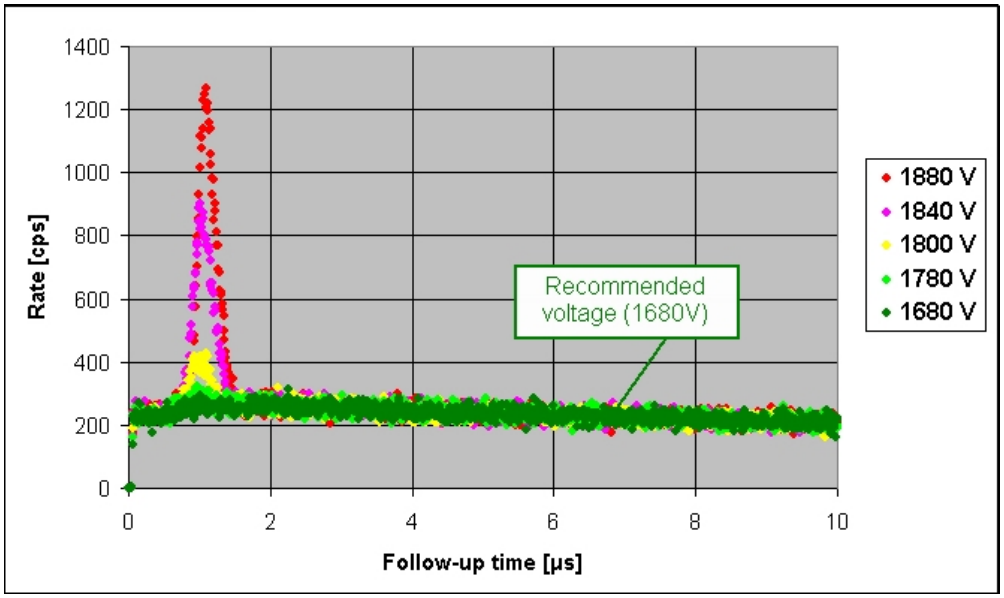

Fig. 5 Follow-up distribution

Presence of excess pulses shows up also in the Rossi-α distribution. The reason for demonstrating it by the follow-up distribution is that calculating Rossi-α distribution is a lengthy operation especially for large impulse trains. In opposite of this displaying follow-up distribution requires no additional calculations, it is even continuously displayed while data acquisition by the PTR system.

4. Functionality testing

PTR system saves channel numbers along with follow-up times. Using channel numbers the impulse rate for each channel can be calculated and displayed. Fig. 6 shows the rate window at a sixteen channel detector with channels #0 to #15 connected. Channel #8 can be identified as defect at once. Also at checking actual rates against calibration values the deviation of a certain channel can be earlier identified when comparing channel values as checking only the overall rate.

Original pulse train can be unfolded into channel files all of which contain data of a single channel. This is made by a simple and quick algorithm; there is no need to repeat the measurement for each channel. In this way all checks can be done on a channel level rather than on the overall impulse train in the same measuring time. Data of an identified unstable channel can be subtracted after the measurement from the impulse train. This may be important when there is no possibility to repeat the measurement e.g. at a safeguard inspection.

Using channel numbers allows also building groups of channels. By this way channel ratio can be measured.

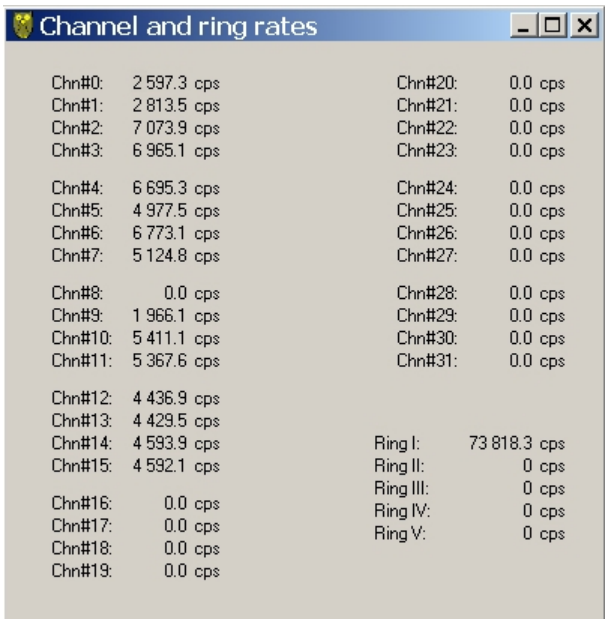

Fig. 6 Channel rates display

5. Die-away time

The HEPC detector designed at PERLA laboratory has an unusual cubic shape [4]. This influences its die-away time. Fitting the usual Rossi-α function with one exponential term gives a relatively good result (Fig. 7). Also the time constant is very close to the one specified by PERLA.

Behaviour of the residuals indicates, however, that the function form is not perfect. Indeed, a fit with two exponential terms produces a perfect fit, but the two time constants (40 and 81 μ) are strongly differing from the original one. Although their weighted average lies pretty near to the original, there is no physical explanation for them. A better approach is to see whether die-away times for the individual channels are differing.

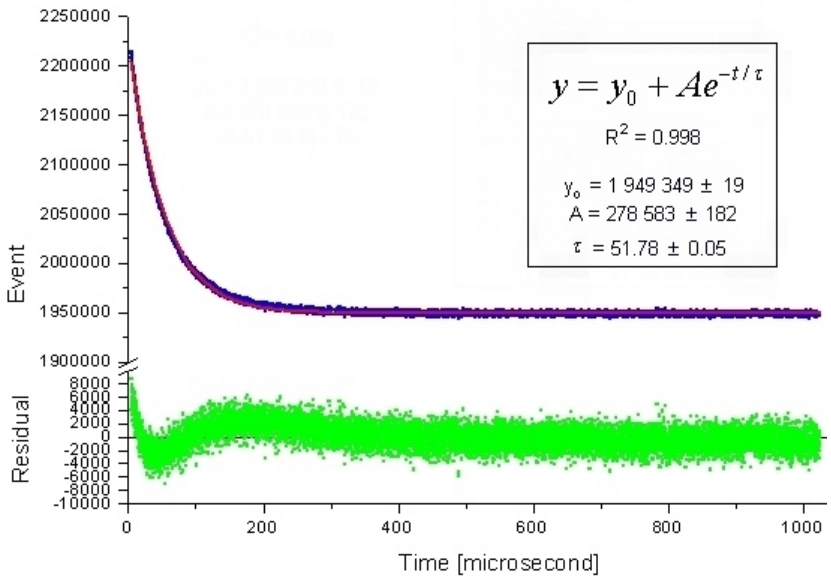

Fig. 7 Overall Rossi-α distribution

For calculating die-away times for single channels impulse trains for individual channels are needed. PTR-32 software can unfold impulse trains as saved data contain also the channel number for each

impulse. There is no need to repeat the measurement for each channel; a simple and quick algorithm unfolds the original file into files all of which contain only data for a single channel.

For the channel files a single exponential term gives also a satisfactory result. Because of the highly reduced impulse rate statistical deviation is increased but residuals show a pretty uniform distribution.

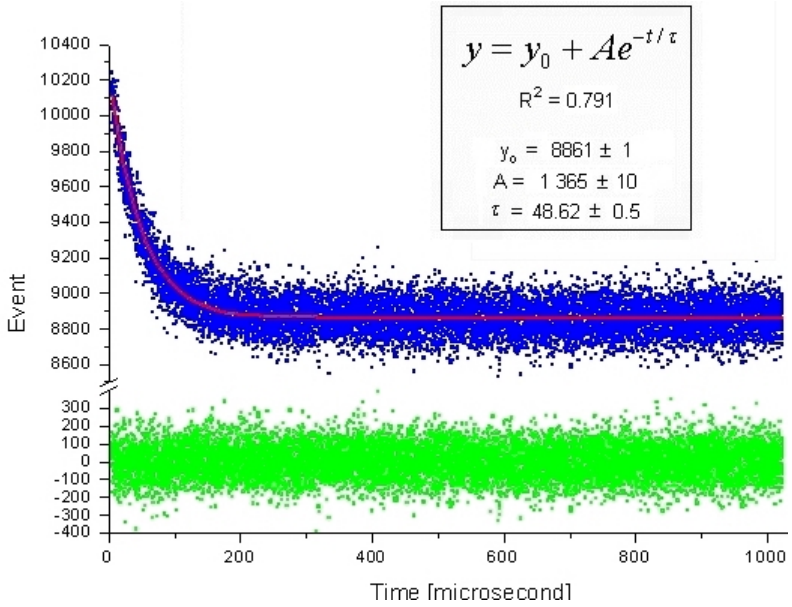

Fig. 8 Single channel Rossi- α fit

Performing the calculation for each channel gives significantly differing die-away times. Results can be separated into three groups around 44 μs, 48 μs and 60 μs. As die-away depends on detector geometry the groups can be assigned to features of the hexahedron. There are 75 detector tubes in the detector connected to 16 preamplifiers. Thus there are 4 to 5 tubes for each amplifier. They are arranged in clusters. Unequal number of detector tubes in clusters and variable distance to the source explain the wide range of measured rates.

Channel		**Rate**	**Tau**
PTR32	**HEPC**	cps	Microsec
0	1A	2 597,33	45,7
1	2A	2 813,51	44,5
2	3A	7 073,98	44,4
3	4A	6 965,12	44,4
4	5A	6 695,36	48,3
5	6A	4 977,54	48,6
6	7AA	6 773,12	47,5
7	1B	5 124,87	48,2
8	7AB	no signal	no signal
9	2B	1 966,13	60,5
10	3B	5 411,18	60,1
11	4B	5 367,65	61,1
12	5B	4 436,90	59,6
13	6B	4 429,57	59,9
14	7BA	4 593,91	58,4
15	7BB	4 592,16	60,4

Table 1 Die-away times

6. Summary

Application of a list-mode pulse train recorder makes the characterization of a neutron coincidence detector easier and quicker since lower number of measurements is required.

The data collected for neutron coincidence counting using the multi-channel PTR contains much more useful information than that recorded with the single-channel version. Die-away time, Rossi-α distribution, single and double count rates can be calculated for individual channels. This can be used to verify the state of health of the detector and allows functionality checking, quality control and optimization channel by channel. The multi-channel list-mode format also has the advantage of removability of noisy channel data. After the modification the data can be revaluated using the same or different Predelay and Gate settings. These features may be beneficial for systems containing a number of detectors.

Necessary hardware is not expensive therefore it could be integrated into detector electronics as an alternative output.

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Perturbation and Burnable Poison Rod corrections for BWR Uranium Neutron Collar

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Abstract:

The active Uranium Neutron Coincidence Collar – Light Water Reactor Fuel (UNCL) provides a means of non-destructively assaying the fissile linear density of fresh fuel assemblies containing low enriched uranium. Historically the generic calibration curves and correction factors were established experimentally on mock assemblies. This is time consuming, expensive and suffers from limitations owing to the limited inventory of pins and simplifications of the laboratory arrangements. At Los Alamos National Laboratory we operate a BWR UNCL-II in conjunction with a 6×6 array of 2.34wt% reference pins. The array is supported by grids held together by steel lifting rods, one at each corner. There is no wrapper. The rods are in interrogation zone. The absence of the wrapper and presence of the support rods constitute a perturbation that requires correction. We have prepared a detailed MCNPX model of the arrangement and computed the correction using the "FT8 CAP tally" correlated neutron capture tally. To establish confidence in our calculations we benchmarked the MCNPX simulations to a series of high precision measurements performed on the mock bundle but two fuel pins replaced by dummy Pb filled pins to change the fuel content and scattering contribution.

Having validated the model we addressed how to construct improved corrections for the presence of Gd_2O_3 burnable poison rods. Experimentally we used 8 poison pins to construct various configurations. These were also modeled to confirm MCNPX could accurately calculate the relative impact.

The MCNPX model will be used to model a variety of enrichments and poison rod configurations covering the anticipated range and arrangement of modern fuel so that suitable correction factors could be estimated. Maintaining traceability to direct physical measurements is essential, but the benefits and flexibility of MCNPX modeling demonstrated in this work for deriving perturbation and poison rod configurations and by extension calibrations for different fuel types is clear.

Keywords: coincidence counter, MCNPX, neutron collar.

1. Introduction

In this paper we describe how the performance of an active Uranium Neutron Coincidence Collar – Light Water Reactor Fuel (UNCL) at Los Alamos National Laboratory has been simulated and we present initial comparisons against experiment as an integral part of validating the model. Neutron collars have long been used routinely by safeguards inspectorates to verify the uranium content in fresh light-water reactor fuel assemblies [1] Both attended and unattended variants have been fielded [2]. The active UNCL is specifically designed to assay the linear density of ^{235}U in fresh fuel assemblies in the axial zone contained by the collar. The principle of the fissile measurement is active coincidence counting whereby the assembly is interrogated by a soft (below the ^{238}U fission threshold) source of random neutrons which induce fission in the assembly. The resulting time correlated pulse train is subject to shift register analysis to extract the rate of coincident pairs known as reals [3]. The bias, deadtime and passive rate corrected reals rate is related to the linear ^{235}U density ($g.cm^{-1}$) through calibration for a particular

assembly design. The ^{238}U linear mass density may also be obtained from analysis of the passive coincidence count data which is dominated by ^{238}U spontaneous fission.

Commercial fuel assemblies come in a wide variety of designs and within a design there are many possible legitimate variants including variation in burn-up and the use of burnable poisons. Assembly designs and loadings have evolved over time as fuel life has been extended; in particular the trend has been to operate to higher burnup which has been made possible through the use of higher initial enrichments and correspondingly higher burnable poison loadings. Historical calibration correlations must therefore be revisited to expand the operational envelope. From the safeguards point of view additionally one must consider a broad range of diversion scenarios and how to resolve problems when the assay result and operator declaration do not agree. Direct calibration against each specific case becomes an impractical proposition and extension from a measured benchmark to an unmeasured configuration cannot be reliably done within the framework of simple analytical scaling rules rooted in the point 'reactor' model. However, detailed Monte Carlo simulation tools such as the Monte Carlo N-Particle Extended code MCNPX ver 2.5.0 or later [4] allows a 3-dimensional representation of the assembly and detector system to be created with high fidelity and the important physical processes of active interrogation and detection to be replicated faithfully, within the limits to which basic nuclear data is known, including a direct shift-register tally capability (the FT8 CAP tally) for a specified predelay and gatewidth. Using MCNPX in this 'complete experimental simulation' way is both conceptually simple and powerful. It overcomes the deficiency of having to break such calculations into separate stages (interrogation and detection) and spatial zones (over each of which the point model can be applied) [12].

More explicitly the calibration curve, that is the reals rate R (pairs per second) as function of ^{235}U linear mass density, m, ($g.cm^{-1}$) is traditionally approximated by:

$$R = k \cdot \frac{a \cdot m}{1 + b \cdot m}$$

where *a* and *b* are empirical calibration factors the scaling factor k is the product of the various correction factor to take into account the difference conditions (detector efficiency, difference in sources, electronics…) from the reference calibration set up. That is [6]:

$$k = k_0 \cdot k_1 \cdot k_2 \cdot k_3 \cdot k_4 \cdot k_5 = \prod_0^5 k_i$$

k_0 accounts for the difference in the Am/Li source strength;
k_1 deals with electronic drift;
k_2 recognizes difference between detector efficiency unit to unit;
k_3 adjusts for burnable poison content (Gd_2O_3 for example);
k_4 corrects for differences in uranium mass per unit length (the so called heavy metal loading $gU.cm^{-1}$);
k_5 incorporates all other effects.

The Monte Carlo method provides a means to evaluate the functional dependence (i.e. the values of *a* and *b*) as well as some of the correction factors. For example k_5, which includes things like the perturbation made by any protective wrappers, is amenable to simulation.

This work represents an ongoing safeguards community wide effort to refine the application of applying computational methods to extend high quality benchmark measurements to new configurations quickly and effectively [see for example [7], and references therein for JRC, Ispra work in this area; and also [8]; [9] for case studies]. Notionally identical collars will show some variation in sensitivity ($reals/sec/g.cm^{-1}$) one to the next even when loaded with the same Am/Li source due to normal (within manufacturing tolerance) dimensional and materials variation and so experimental benchmarking (ideally to a common reference condition for a given instrument type) cannot be avoided altogether when high accuracy is needed, but transferring a calibration is based on ratios of responses and is far less variable across an instrument type. A detailed description of the calibration procedure currently used by the safeguards inspectorates may be found in LA-11965-MS [10]. Note that the neutron output of the MRC-95 Am/Li source was subsequently revised downward [11,12] as we shall explain later.

2. UNCL Design

The UNCL collar was of the prototype UNCL-II design (units LANL-3, LANL-4) introduced in 1989 [Menlove et al, 1990] (nominal CANBERRA Industries model JCC-73) in the BWR configuration but with upgraded electronics compared to what was originally used. There is no Cd anywhere in the system.

The collar consists of a high density polyethylene (HDPE) body of rather intricate shape. The internal cavity is notionally 16.5 cm across. The bottom of the collar is an Al-plate 0.65 cm thick that covers the void space left under the ^{3}He proportional counters. This rests on a cart so that the base was modeled at 10 mm thick. The underside of the base is approximately 65.9 cm above the concrete floor. On top of the collar is a junction box containing the high voltage and low voltage planes and three Amptek A-111 based amplifier-discriminator units. The junction box is machined from Al. The inners are not modeled and the box is notionally represented by a 1cm plate of Al. The junction box is in a region of relatively low neutron importance and so detail modeling is not required.

The collar contains a total of 16 ^{3}He proportional counters arranged in the two side walls and the rear wall (opposite the Am/Li source). The tubes are of the RS-P4-0813-101 type. The tube wall is made of 1100-F aluminum 0.508mm thick. The outer diameter is 25.4 mm with a maximum tolerance of 0.762 mm (0.03"). The nominal active length is (330.30±0.76) mm (13"±0.03") as defined by the distance between the anode supports and commences (23.876±0.508) mm from the external base rum of the tube. The tubes are filled to a partial pressure of 4 atm in ^{3}He. 1 atm = 101.325 kPa at 760 mmHg and 0ºC but in this case referred to 25 ºC corresponding to a He number density of 9.845906×10^{-5} atom.(b.cm)$^{-1}$ based on the ideal gas laws. It is reasonable to take the He-atom number density to be equal to the ^{3}He number density because ^{3}He gas is typically >99.98 ^{3}He by atom %. Details of the connector and other ^{3}He tube features such as the anode wire are not included in the MCNPX model since they have a trivial influence.

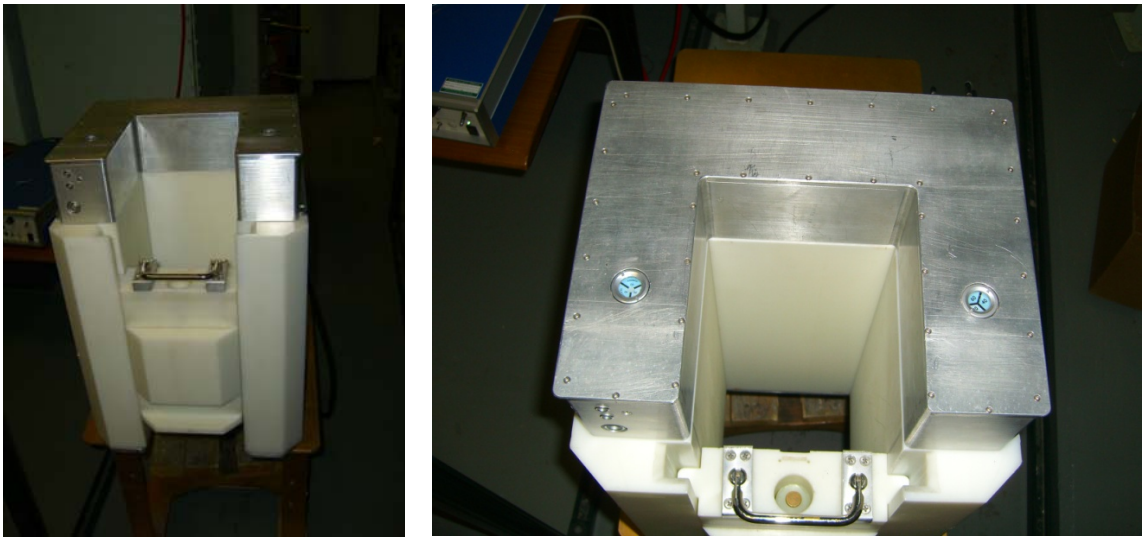


Figure 1: Two photographs of the UNCL-II used at LANL to generate these results

3. Am/Li Source

For absolute comparison the yield of the ^{241}Am/Li source is needed. Our Am/Li source MRC-95 was chosen long ago as the working standard to which others are referred [10]. In that report the yield is quoted as 3.96×10^{4} n.s^{-1} in Jan 1989 although later re-determination resulted in a revision downward to $(3.349 \pm 0.015) \times 10^{4}$ n.s^{-1} [11,12] referred to May 17th 1999 (although with a few weeks the exact date is

unimportant for decay corrections given the long half-life of ^{241}Am). A formal LANL memorandum, NIS-5-99-152, summarizing the revision was also prepared by H. Menlove, D Mayo and P. Rinard. Titled 'Absolute calibration and energy spectrum of AmLi neutron sources' the memorandum is dated May 17th 1999. The active volume was taken to be a cylinder 2cm in diameter by 3 cm long containing approximately approximately 0.2 g of ^{241}Am (~0.68 Ci) in the form of dioxide mixed with approximately 2.3 g of Li. The active source is doubly encapsulated in 304-type stainless steel. To reduce the gamma-ray dose from ^{241}Am the source capsule is contained in a screw top tungsten container with a minimum wall thickness of 0.25 cm. A threaded hole mates the tungsten container to a polyethylene rod which serves as a carrying device and also reflector when the source-assembly is placed into the receiving hole of the lift-out 'door' of the UNCL when an active assay is in progress.

In this work source MRC-117 was used. This has an output 1.220 times greater than MRC-95 [10]. The neutron emission spectrum was taken from the SOURCES-4A code [12].

4. Fuel Bundle Description

The 36 BWR fuel rods used in this work were supplied to Los Alamos by United Nuclear Corporation Research and engineering Center in 1971. The rods contained UO_2 ceramic pellets of density 10.48 g.cm^{-3} and diameter 1.224 cm (0.482"). The enrichment is 2.34 wt% in ^{235}U as a fraction of total U. Each rod contains nominally the same mass of fuel, 1504 g of UO_2. The length if the stack was given as 121.9 cm (48.0") by the manufacturer and this was confirmed within limits (122.5±0.5) cm by a gamma scan using a HPGe gamma-ray spectrometer fitted with a narrow slot collimator. The full rod length is 130.0 cm. The cladding is Zircaloy-2 0.089 cm wall thickness (0.035") with an outer diameter of 1.429 cm (0.5625"). The rods are held in a 6x6 array on a 1.9 cm pitch by a pair of Al grid plates above and below the region of active interrogation and outside the physical extent of the collar so that they exert negligible influence on the observed response. The grids are held in place by four steel tie/lifting bars, one passing through each corner of the grid plates. The bars are 1.43 cm in diameter and form a square 13.8 cm between centers.

The fuel pins may be replaced by Pb dummies of the same external dimensions and clad as the actual rods. In the case that a poison rod is to be substituted the 12 PWR poison rods are available. These contain UO_2 pellets of 10.48 g.cm^{-1} enriched to 3.27wt% in ^{235}U. The pellets are ~0.90765 cm in diameter and form a stack length of 104.0 cm in a rod of overall length 130.0 cm. The rods have an outer diameter of 1.08 cm with the Ziraloy-4 clad being 0.115 cm thick. There is 571 g of total U per rod and 40.6 g of Gd_2O_3. Homogenized this equates to a Gd_2O_3+UO_2 matrix of effective density 10.23 g.cm^{-3} [13].

5. Experimental Activities

The instrument was set up in the basement of Blg27 at Los Alamos according to recommended operating parameters, namely a predelay of 4.5 μs, a coincidence gatewidth of 64μs and a high voltage of 1680 V. Deadtime corrections are modest at the rates observed and in the case of the Real coincidence rate the correction factor traditionally calculated as $\exp[(A+B.T).T]$ where T is the observed totals rate and A and B are previously determined reals dead time parameters is normally NOT made, that is to say A is set to 0.0 x10^{-6} s and B (~A^2/4) is set to 0.0 x10^{-12} s^2 for BOTH calibration and assay. Since the totals rate typically is <<1/A and does not vary much from assembly to assembly this is a valid simplification.

For convenience in the present measurements the fuel bundle is positioned symmetrically left to right inside the collar but with the steel tie bars touching the inner face of the lift-out 'door' which houses the Am/Li source. This ensures easy reproducibility of the geometry.

In the field a typical assay may comprise a 200 sec passive count followed by a 600 sec active count. The statistical uncertainty on the net reals rate for the 6x6 pin array is of the order of ±1.3% in the present

configuration. This is adequate for initial scoping studies but in future work we'd like to get sub 1% precision and so count times will be extended to 600 sec (20x30 sec) and 3600 sec (60x60 sec) as a minimum which should result in a precision of ~0.6% based on inverse root time scaling.

The LANL-4 UNCL-II unit has just a single combined signal output so experimentally we can not attempt to locate substitutions or poison pins but in MCNPX we can tally the hit pattern amongst the ^{3}He tubes and ask whether it would in principle be possible in which case the future prospect of using one amplifier per tubes and list mode data acquisition could be considered as an upgrade route.

6. Model Construction

The MCNPX input was created independently but benefited from extensive prior art in our group against which aspect could be checked [13]. In MCNPX often a given description can be coded in various ways and different users have their own preferences and style. So having this independent check is invaluable for catching mistakes. In our case we found an error in the work cited of the specification of the ^{3}He tube diameter and wall thickness. Visualization of the model is another powerful aid. We took data from reports, such as delivery notes and blueprints, but where possible we confirmed this direct observation and dimensional measurements on the system. The model is more geometrically more detailed than would normally be the case in the community and reflects the increased computer speed available meaning that simplifications and approximations no longer need to be as severe. There is some penalty in set-up time however. Finally independent peer review was applied to the input decks.

7. Monte Carlo Numerical Results

The Monte Carlo numerical calculations were performed using MCNPX 2.7c LANL code. Figure 2 shows the cross section of the collar as introduced in the code; the UNCL-II surrounds the middle of the BWR fuel assembly. In all our simulations we have set up analog capture, FT8 CAP tally with 4.5 μs pre-delay and 64 μs coincidence gate, and ran for 10^{7} histories (that is (α,n) neutrons launched). The run times were approximately 1 hour in duration sufficient to obtain a standard deviation of less than or about 1%. Subsequently we repeated the runs on a cluster of processors to substantially improve on the MCNPX sampling uncertainty (but these results will be reported elsewhere). It is worth mentioning that in the input decks we have also modeled the room to consider in the simulations radiation scattering.

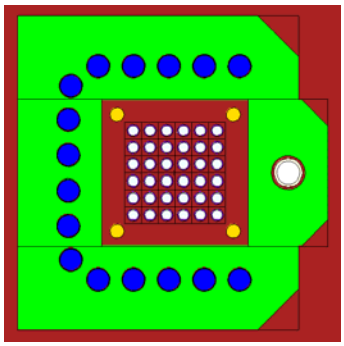
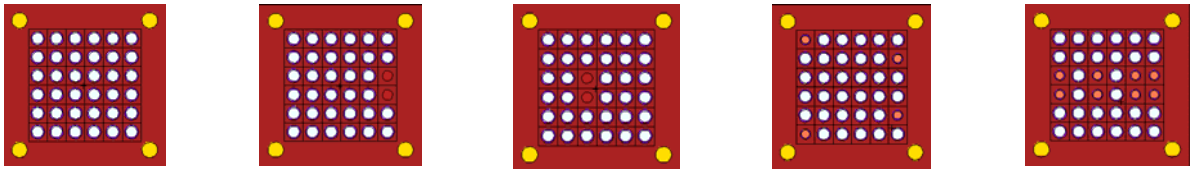


Figure 2: UNCL-II cross-sectional view plotted from the MNCPX input file. In blue are visible the ^{3}He proportional counters, in green the HDPE body, in red the air, and in orange the fuel grid steel tie-bar supports. In the center may be seen the 6x6 fuel pin array.

To initially benchmark the modeling against real measurements we report here just the following five scenarios (see Figure 3): in one case all of the array positions were filled with BWR fuel pins (enrichment 2.34wt%), in two cases two fuel pins were replaced with 2 dummy rods containing lead (see Figure 3 for

the positions), in another case 4 poisons rods were used to replaced fuel pins, and in the fifth case 8 poisons rods replaced fuel pins. The results of the Monte Carlo simulations are presented in Table 1 in comparison with the measurements. Simulations and measurements are in very good agreement, in the context of the experimental precision, as is indicated by the Delta Double column in the Table 1.



Case 1: full | Case 2: 2 dummies | Case 3: 2 dummies | Case 4: 4 poison rods | Case 5: 8 poison rods

Figure 3: Five scenarios both modeled in Monte Carlo and measured in laboratory

CASES	Doubles Rate (Measured)	Doubles Rate (Calculated)	Delta Double (%)
Case 1: full	150.7 (1.3%)	148.5	-1.5
Case 2: 2 dummies	143.3 (1.3%)	142.7	-0.4
Case 3: 2 dummies	146.2 (1.3%)	142.9	-2.3
Case 4: 4 poison rods	124.5 (1.5%)	121.3	-2.6
Case 5: 8 poison rods	90.0 (1.8%)	84.7	-5.9

Table 1: Results of the Monte Carlo simulations in comparison with measurements. The uncertainties reported in brackets for the measurements are 1σ relative standard deviations (68.3% confidence limits). Corresponding sampling uncertainties on the calculated values are less than 1%.

It is important to emphasize that the comparisons in Table 1 are on an absolute basis. Relative predictions give some canceling of bias.

As a perturbation case of interest we also considered the effect of removing the steel support bars: for this case Monte Carlo calculations give a correction factor of 1.05 (larger than the factor of 1.03 historically adopted). Experimental comparisons are pending.

8. Future Work

No physical measurement or scientific calculation is meaningful without an accompanying statement of uncertainty or confidence and a description of how the uncertainty was estimated and how it relates to other quantities and result – the covariance structure. The uncertainty evaluation should be realistic, justifiable and fit for purpose. This is because the quality of subsequent use depends strongly on the accuracy and correct interpretation of the input. Often uncertainties are under reported and poorly described. This is a well known problem in the specialism of nuclear data evaluation [14]. In discussing how the data evaluator may deal with and resolve discrepancies the reader of Peelle's paper is also reminded of common pitfalls and how to avoid them. The thoughtful and careful worker should therefore take pains to identify all important influences, quantify their effects and document what was done. In the present context we recognize that the results presented are preliminary and lacking the detailed sensitivity analysis required place an uncertainty band on the results – for example sensitivity to the specification of the Am/Li spectrum or of the effective active length of the ^{3}He proportional counters or of the HDPE density, or of the choice of cross sections etc.. It is intended that the uncertainty budget will be

addressed in detail in future work although we are certainly encouraged by the initial agreement obtained. As an example of fine tuning at the fraction of a per cent level consider the that in the simulations every ^{3}He(n,p) reaction is assumed to result in a detected count whereas in practice wall and end effects could lead to losses and ions from notionally dead space could enhance that rate a little. The fact that not all events are counted is witnessed by the small but finite slope of the high voltage plateau. Given the capability of simulation tools such as MCNPX to replicate the neutron transport problem so well, fine details such as this, and which don't impact ratio calculations, must be addressed if we are to push back the state of the art of absolute performance prediction.

The experimental data will be extended and precision improved so that a more stringent test of the MCNPX model can be undertaken. Progress was hampered while a criticality safety review was being undertaken but we look forward to taking fresh data including results for triples counting. We also intend to apply totals-based algorithms to strengthen the safeguards conclusion from the collar for instance as has been proposed for the case of the active well coincidence counter [15]. Our hope is to exploit the active singles count to glean information on the poisons independent of the declaration. This would give a more powerful assay tool, one less reliant on the operator.

Many other comparisons are possible but which we did not discuss here. These include passive rates, rates with the Am/Li source loaded but no fuel rods present, dieaway profile, ^{252}Cf efficiency scans etc. These are reserved for a future report. It should be borne in mind too that variation from instrument to instrument is to be expected due to normal fabrication tolerances and component variability. From Tables XII-XV in Menlove et al 1990 [10] it would seem that a variation of few % in the case of the four UNCL-II units studied is to be expected. Tracy R Wenz of LANL IAT-1 has also provided the authors with unpublished collar inter-comparison data which supports this. Better absolute agreement than this with the simulation results for a particular instrument may therefore be considered fortuitous.

9. Conclusions

Motivated by the need to extend the commonly adopted active neutron coincidence collar calibration correlations for fresh light water reactor fuel elements to higher initial enrichments and higher gadolinia content we have created a new MCNPX model of the LANL-4 UNCL-II instrument. Preliminary results have been presented and are seen to be encouraging. A justifiable perturbation factor for the steel tie bars (1.05) has been calculated to replace the historically used correction factor (1.03) which is part of k_5. Replacement studies have shown that the relative response of different configurations can be calculated faithfully. This gives us confidence in the basic correctness of the model. The next step is to run many more cases and compare them with high quality measurements to test the limitations of the predictive capability. Trend for modern fuels will then be calculated and appropriate uncertainty bounds evaluated.

10. Acknowledgements

This work is supported by the U.S. DOE and in particular is a foundation element to Action Sheet 14 between The National Nuclear Energy Commission of Brazil (CNEN) and the United States Department of Energy (DOE).

We also wish to thank Michael C. Browne for encouragement and support throughout on this project and Carlos D. Rael for practical help.

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Calibration and Monte Carlo Modelling of a fast-UNCL for the IAEA

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Abstract

A campaign of measurements has been carried in the JRC PERLA laboratories in order to calibrate and characterise a fast-UNCL counter recently acquired by the IAEA. Various radionuclide neutron sources (^{252}Cf, ^{241}AmLi), BWR and PWR reference assemblies were used. Furthermore, the counter has been extensively modelled using the Monte Carlo code, MCNP-PTA, which simulates both the neutron transport and the coincidence electronics. The models have been validated using measurements, the results of which agree generally very well with the simulations. The WWER1000 fuel magazine for which there are no representative reference materials for an adequate classical calibration of the counter, has also been modelled using MCNP-PTA. The response of the fast-UNCL to these fuel assemblies has been simulated and numerical calibrations curves were obtained for the above fuel assemblies in various modes (fast and thermal). They are compared to a standard UNCL-II from LANL. Preliminary results are given as further work including, sensitivity analysis, model validation for WWER10000 fuel, efficiency optimization for BWR etc. is in progress.

Keywords: Neutron counting, Calibration, NDA, Monte Carlo simulation

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Introduction

The Antech 2044 collar [1] hereafter referred to as fast-UNCL (or F-UNCL) is an active NDA system built by Antech (UK) for IAEA to be used for the determination of the ^{235}U content in fresh fuel assemblies. In its active configuration, (one of the PE slabs containing an ^{241}Am-li(n,α)) the F-UNCL has 37 ^{3}He tubes at 10 atm. of pressure compared to 18 tubes at 4 atm. of pressure for a standard collar. To be used in a passive configuration, the F-UNCL polyethylene slab with an ^{241}Am-li(n,α) is replaced with a slab which houses an additional nine ^{3}He tubes to make a total of 44. Furthermore the internal cavity of the UNCL is adjustable to efficiently house the variety of Fuel assemblies as intended.

Figure 1: A photo of the F-UNCL in its passive configuration

The IAEA shipped the F-UNCL to the Perla laboratories belonging the JRC-ITU Security Unit (Ispra) where work was undertaken in order to:

1) Characterise and calibrate the counter using measurements with calibrated radionuclide sources and reference materials including ^{252}Cf and ^{241}AmLi as well as BWR and PWR fuel reference.
2) The response or efficiency of the F-UNCL to the above sources of neurons as well as the operating parameters of the collar would subsequently be determined together with its rho-zero.
3) Develop a Monte Carlo model of the F-UNCL for both the active and passive configurations, both in fast and thermal modes using MCNP-PTA [2-4].
4) Validate the model by comparing measurements and simulations for various configurations and neutron sources, including BWR, PWR fuels which were also simulated together with the WWER1000 fuel although no measurement data is available at the present time of the latter fuel.
5) Once the model is validated, calibration curves were developed for BWR, PWR, WWER1000 fuel assembly both in fast and thermal modes.

In this paper we describe the work carried out in this context and present a summary of the preliminary results that our measurements and calculations have yielded. Furthermore, comparison with the UNCL-II of LANL [5] has been undertaken and will be presented here. This work was performed at the ITU-Nuclear Security Unit (Ispra) under the EC Support Programme to IAEA Task EC-A-1505 (Numerical Calibration of Neutron Coincidence Counting Instruments).

1 Characterisation of the fast-UNCL in PERLA

The characterisation and the determination of the main operational parameters of the 2044 collar were performed in the PERLA laboratory. It included the following measurements, determination of optimal high voltage, detection efficiency at the centre of the cavity, dead-time correction factors, dye-away time, efficiency profiles, response of the counter to reference radioactive sources, PuGa standards and various fuel elements such PWR , BWR and WWER and finally the determination of rho-zero of the counter. The details of the characterisation campaign are given in Table 1 which summarises the most important parameters and characteristics of the counter.

Optimal high voltage	1750 V
Detection efficiency for ^{252}Cf at the centre of the cavity:	
- Passive configuration without Cd	0.171±0.002
- Passive configuration with Cd (0.35mm)	0.156±0.002
- Passive configuration with Cd (0.5mm)	0.156±0.002
- Active configuration without Cd	0.144±0.002
- Active configuration with Cd (0.35mm)	0.128±0.002
Dead-time correction factors:	
- Passive configuration	a: $0.79*10^{-6}$ s b: $4.4*10^{-12}$ s^2
- Active configuration	a: $1.12*10^{-6}$ s b: $3.8*10^{-12}$ s^2
Die-away time:	
- Passive configuration without Cd	38.0 μs
- Passive configuration with Cd (0.35mm)	33.4 μs
- Active configuration without Cd	41.2 μs
- Active configuration with Cd (0.35mm)	33.2 μs
Rho-zero	0.096

Table 1: Main operational parameters of the 2044 collar deduced from measurements

2 Monte Carlo Modelling of the counter and Validation Measurements

The following sections aim to describe the Monte Carlo modelling of the F-UNCL and its validation. All calculations have been done using the MCNP-PTA code [3,4] and compared to measurements for validation.

2.1 Response to ^{252}Cf source spectra

The response of the F-UNCL to a ^{252}Cf neutron source (number NC6005) with and without Cadmium in the Passive Configuration was calculated using MCNP-PTA and compared to measurements (Background is subtracted). As seen in Tables 2 and 3, an excellent agreement is obtained between measurements and Monte Carlo calculations. The Cf-252 (6005) with a neutron yield of $1.623*10^3$ n/s (on 17.05.2010) was centrally positioned, 22 cm from the bottom the counter, and the counter operated at 1750 V HV, a pre-delay of 4 µs and Gate width of 64 µs.

Cd (mm)	S (cps)		D (cps)		T (cps)		Efficiency	
	Measured	MCNP	Measured	MCNP	Measured	MCNP	Measured	MCNP
0	280±1	273.8±0.1	47.9±0.3	48.0±0.1	4.9±0.1	4.8±0.1	0.171±0.002	0.168±0.002
0.35	256±1	250.0±0.1	44.4±0.3	44.3±0.1	4.3±0.1	4.2±0.1	0.156±0.002	0.152±0.002
0.5	256±1	249.5±0.1	44.4±0.3	44.1±0.1	4.3±0.1	4.2±0.1	0.156±0.002	0.152±0.002

Table 2: Results of Calculations compared to measurements (Background is subtracted) for a ^{252}Cf source (no. 6005) with and without Cd with the F-UNCL in the Passive Configuration

Cd mm	S (cps)		D (cps)		T (cps)		Detection Efficiency	
	Measured	MCNP	Measured	MCNP	Measured	MCNP	Measured	MCNP
0	234.0±0.5	229.1±0.1	31.7±0.3	31.8±0.1	2.7±0.1	2.6±0.1	0.144±0.002	0.142±0.003
0.35	206.7±0.3	203.0±0.1	28.7±0.2	28.9±0.1	2.2±0.1	2.3±0.1	0.128±0.002	0.126±0.003
0.5	--	202.0±0.1	--	28.8±0.1	--	2.3±0.1	--	0.126±0.003

Table 3: Results of calculations compared to measurements (Background is subtracted) for a ^{252}Cf source (no. 6005) centrally positioned with and without Cd in the active Configuration (i.e. front panel changed to contain only the AmLi source hole and no AmLi or ^{3}He tubes).

2.2 Response to Reference PWR310 fuel assembly

The PERLA PWR reference fuel element was measured in active configuration using the certified Am-Li source ANHP-N008. This source has an intensity certified by NPL of 59660±840 n/s at the reference date of 19th March 1999. The Perla PWR reference fuel element is a 17x17 assembly containing 264 fuel rods. Each rod has a length of 129.59 cm and is filled with 3.1% enriched UO_2 pellets. Its linear fissile content is 39.90 g^{235}U/cm and the U loading linear mass is 1271.5 (g/cm). The PWR assembly was positioned at the centre of the cavity (Figure 2). Table 4 collects all the measurements performed with the PWR reference fuel assembly. The response of the F-UNCL to reference PWR fuel assembly available in the PERLA laboratories was also calculated using MCNP-

PTA and compared to measurements with and without cadmium, the results of which are shown in Table 4 with a good agreement is again obtained between measurements and calculations. The parameters used in both calculations and measurements are the following: pre-delay = 4.0 μs, gate = 64 μs, deadtime=1.12 μs, pulsetime = 50.ns, DT= 1.12 μs in active and 0.79 μs in passive mode.

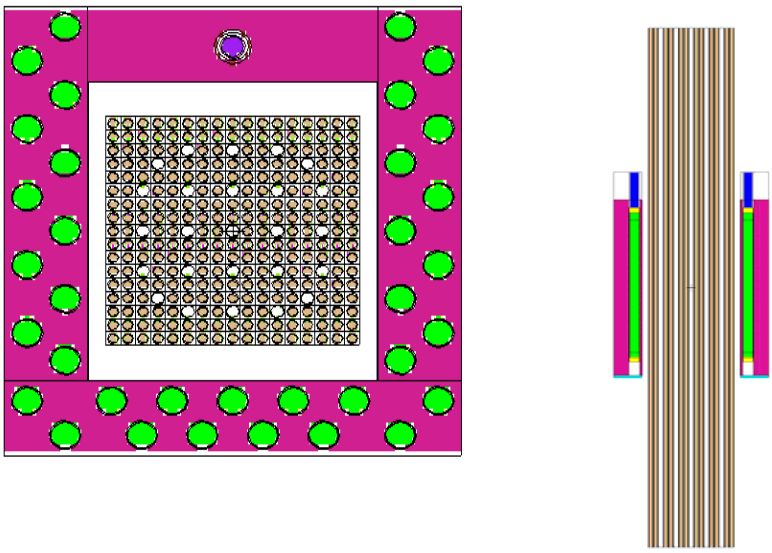

Figure 2: MCNP Model of F-UNCL in Active configuration with a PWR fuel assembly in place.

Source	Cd (mm)	S (cps)		D (cps)		T (cps)	
		Measured	MCNP	Measured	MCNP	Measured	MCNP
Active backgrnd AmLi 008	0	6437.4 ±0.3	6928.9±0.6	-0.2 ±0.4	-0.63 ±0.60	0.06 ±0.13	-0.08 ±0.3
Active backgrd AmLi 008	0.35	3921.0 ±0.2	4269.0 ±0.5	0.28 ±0.26	0.29± 0.38	0.16 ±0.04	0.02±0.14
PWR310 No AmLi	0	196.2 ±0.3	170.7 ± 0.1	19.43 ±0.03	19.22±0.01	2.2 ±0.1	1.86±0.01
PWR310+AmLi008	0	6676.9 ±0.8	6705.6±0.6	397.5 ±0.8	401.6±0.6	41.1 ±0.5	41.98±0.36
PWR No AmLi	0.35	165.0 ±0.2	143.3 ± 0.1	13.3 ±0.2	13.3 ±0.1	1.35 ±0.05	0.98 ±0.10
PWR310+AmLi008	0.35	3468.1 ±1.1	3444.1 ± 0.4	49.7 ±0.8	36.7± 0.3	4.6 ±0.3	2.97±0.10

Table 4: Response of the F-UNCL in the thermal and passive configuration to reference PWR fuel elements calculated by MCNP-PTA and compared to measurements .

2.3 Response to Reference BWR fuel assembly

The Perla BWR reference fuel assembly consists in an 9x9 assembly of rods. Differently from the PWR assembly the 9 by 9 BWR assembly contain a mixture of 21 rods at 1% enrichment in ^{235}U, 36 rods at 3.1% and 20 rods at 5% which correspond to a linear fissile content of 3.9491, 12.2421 and 19.7454 g ^{235}U/cm, respectively. The U loading linear mass is 366 g/cm. Whereas the PWR assembly was centrally placed, the BWR assembly was positioned eccentric to the cavity at approximately 1.5

cm from the source-holder slab. The response of the F-UNCL to reference BWR fuel assembly was also calculated using MCNP-PTA and compared to measurements with and without cadmium, as shown in Table 5. A good agreement is again obtained between measurements and calculations. The parameters used in both calculations and measurements are the same as above for the PWR.

Source - mode	Cd (mm)	S (cps)		D (cps)		T (cps)	
		Measured	MCNP	Measured	MCNP	Measured	MCNP
Backgrnd Active	0	6437.4 ±0.3	6928.9±0.6	-0.2 ±0.4	-0.63 ±0.60	0.06 ±0.13	-0.08 ±0.3
Backgrd Active	0.35	3921.0 ±0.2	4269.0±0.5	0.28 ±0.26	0.29± 0.38	0.16 ±0.04	0.02±0.14
BWR310 Passive	0	55.8 ±0.2	42.3±0.1	2.95±0.04	3.08± 0.01	0.18 ±0.01	0.17 ±0.01
BWR310, Active	0	6340.6 ±1.6	6779.1±0.5	156.3 ±1.5	167.7± 0.4	9.8 ±0.8	9.94±0.21
BWR310 Passive	0.35	47.8 ±0.2	35.8±0.1	2.23 ±0.24	2.40±0.01	0.13 ±0.01	0.11±0.01
BWR310 Active	0.35	3522.8 ±0.8	3787.8±0.5	15.4 ±0.6	11.81±0.30	0.9 ±0.2	0.59±0.12

Table 5: Response of the F-UNCL ,in the thermal and fast modes and for both the active (AmLi) and passive configurations, to reference BWR fuel elements calculated by MCNP-PTA and compared to measurements.

2.4 Response to wwer1000 fuel assembly

The response of the F-UNCL with and without cadmium to reference WWER1000 fuel assembly, the MCNP model of which is shown in Figure 3, were calculated and results are shown in Table 6. The parameters used in the calculations are the same as for PWR and BWR. Measurement data would be obtained in due time and compared to calculations.

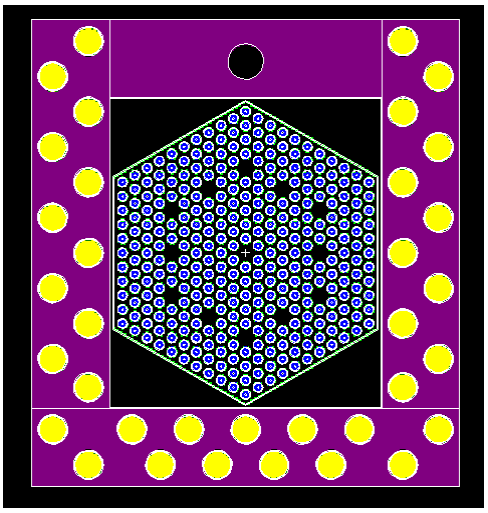

Figure 3: Model of F-UNCL in Active configuration with a WWER1000 fuel in place

Fuel / Mode	Cd (mm)	S (cps)	D (cps)	T (cps)
wwer1000 Active	0	7557.0 ± 1.8	506.8 ± 1.6	58.9 ± 1.2
wwer1000 Passive	0.35	3814.8 ± 1.3	54.39±0.81	4.70±0.41
wwer1000 Passive	0.5	3829.6 ± 1.3	55.29 ± 1.0	4.89 ± 0.46

Table 6: MCNP-PTA calculated response of the F-UNCL to WWER1000 fuel elements in various modes and configurations .

2.3 Numerical Calibration of the F-UNCL for PWR, BWR and WWER1000

MCNP-PTA was run for a range of ^{235}U enrichment (natural to 5%) of the PWR, BWR and WWER1000 fuel assemblies, both in fast and in thermal modes (with and without 0.35 mm thick Cadmium).

Using the DEMING code, both the fast and thermal mode data are fitted by a function of the form:

$$y = (a * x) / (1 + b * x)$$ where x = linear ^{235}U mass in g/cm and y = doubles rate

resulting in parameters given in Table 7 which also give parameters of calibration curves obtained in a similar way for BWR and WWER1000.

Fuel, Mode	**Model**	**a** ±	**b** ±
PWR, thermal	a.x/(1+b.x)	25.725 ± 0.382	0.038±0.0009
PWR, fast (Cd)	a.x/(1+b.x)	1.074 ± 1.657E-2	3.457E-3 ± 3.819E-4
BWR, thermal	a.x/(1+b.x)	30.001 ± 2.632E-1	8.903E-2 ± 1.528E-3
BWR, fast	a.x/(1+b.x)	1.2449 ± 3.307E-02	1.334E-2 ± 2.307E-3
WWER1000, thermal	a.x/(1+b.x)	27.463±0.537	0.0404 ± 0.0013
WWER100, fast	a.x/(1+b.x)	1.168 ± 0.018	5.409E-3 ± 4.156E-4

Table 7 : Summary calibration parameters of the F-UNCL for the three fuel assemblies and in fast (with Cd) and thermal (no Cd) configurations.

5. Comparison of FUNCL response to that of UNCL-II collar of LANL

5.1 PWR configuration – thermal and fast modes

All the measurements in PERLA were done using the AmLi source ANHP-N008 whereas MRC95 was used by LANL [2] as a reference, as given in Table 8 for comparison. The PERLA-PWR standard which is a 264-rod assembly with 17x17 array of pins containing 3.1% enriched UO_2 fuel is compared to LANL reference assembly as shown in Table 9. As written above, the Perla BWR reference fuel assembly is a 9x9 assembly, containing 77 of rods made of a mixture of 21 rods at 1% enrichment in ^{235}U, 36 rods at 3.1% and 20 rods at 5% which, correspond to a linear fissile content of 3.9491, 12.2421 and 19.7454 g ^{235}U/cm respectively. The U loading linear mass is 366 g/cm.

	MRC95	N800
Reference date	1/1/1989	19/3/1999
Fluence on ref date	39600 n/s	59660
Fluence in May 2010	38263	58600

Table 8: Details of Am-Li sources

	PERLA PWR	PERLA BWR	LANL PWR	LANL BWR
Active length (cm)	129.6	129.6	103.5	103.5
^{235}U/U abundance wt %	3.11	1,3.1,5	3.19	3.19
Rod lattice	17 x 17	9x9	15 x 15	9 x 9
Nr. of fuel rods	264	77	204	76
Rod pitch (cm)	1.257	1.257		
Cladding outer diameter (cm)	0.968	0.968	0.968	10.48
Cladding inner diameter (cm)	0.854	0.854	0.854	
Pellet diameter (cm)	0.8123	0.8123	0.8123	
UO_2 fuel density (g/cm^3)	10.46	10.46	10.48	10.48
U-235 loading mass (g/cm)	38.99	11.979	38.76	14.44
U loading mass (g/cm)	1261.5	366	1215	453

Table 9: Details of the PWR, BWR reference assemblies of PERLA and LANL

From the assembly characteristics given above and following the methodology detailed by Menlove et al. [5] one can compute the uranium mass loading k_4 factor [5] for the PERLA (PWR or BWR) assemblies and be able to relate the LANL UNCL reference calibration curves [5] to those of fast-UNCL, the parameters of which were given in table 7 above.

A comparison of the response of the F-UNCL with LANL standard to PWR and BWR (fast and thermal modes) using the MRC95 AmLI source within the UNCL-II collar was thus made. The difference in amplitude of Reals reflects the efficiency of the counters.

Noteworthy, two AmLi sources are involved here, the N008 to MRC95, whose ratio of yields deduced from the nominal values given by the manufacturers equals 1.52 results. However, a ratio equal to 1.72 was deduced in reference [6] from 3 sources (C282, C299, C300) which were measured both in Perla and at the LANL and their ratios reported.

Table 10 gives the ratio of the response of the IAEA fast-UNCL to that of the LANL UNCL-II, for fast and thermal modes, to PWR and BWR fuel assemblies for 3 different % of U235 enrichments. Values are given for source normalization factors equal 1.72.

	Fast PWR	Thermal PWR	Fast BWR	Thermal BWR
0.7% ^{235}U	1.45	1.40	0.88	0.68
3.1% ^{235}U	1.57	1.23	0.91	0.79
5% ^{235}U	1.65	1.18	0.93	0.84

Table 10: Ratio of the response of the IAEA fast-UNCL to that of UNCL-II of LANL in fast and thermal modes and for PWR and BWR fuel assemblies with 3 different % of U235 enrichments (source normalization ratio 1.72)

Summary and conclusions:

Preliminary results of measurements carried out in PERLA laboratories using the IAEA Fast-UNCL counter and a variety of calibrated radionuclide sources and reference materials including ^{252}Cf and ^{241}AmLi, as well as BWR and PWR fuel reference were given here. The F-UNCL has thus been fully characterised and calibrated and the response or efficiency of the F-UNCL as well as the operating parameters of the collar were subsequently determined from both measurements and calculations. Monte Carlo models of the F-UNCL for both the active and passive configurations, in fast and thermal modes were developed and the detector's responses were determined. The models were validated by comparing measurements and simulations for various configurations and neutron sources, including BWR, PWR fuels. These fuel assemblies, including the WWER1000 fuel for which no measurement data is available at the present time, were simulated and good agreement between measurements and Monte Carlo calculations was generally obtained. Subsequent to model validation, calibration curves were developed for BWR, PWR and WWER1000 fuel assemblies both in fast and thermal modes and the responses of the F-UNCL to BWR and PWR (in thermal and fast modes) were compared to those of the reference UNCL-II counter from LANL. Further work, including the optimisation of various operational parameters, efficiency optimization for BWR, sensitivity analysis of the counter, model validation for WWER10000 fuel etc. is in progress.

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16 Non-proliferation: experience and future challenges

The Impact of Interdependencies on the Effectiveness and Efficiency of Safeguards

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Abstract:

The purpose of Integrated Safeguards is to evaluate a State as a whole and provide credible assurance of the absence of undeclared nuclear activities. The new system is not viewed as an addition to the traditional safeguards system but as a collection of measures to verify the correctness and completeness of the declarations.

The broadening of the scope to evaluate the State for the purpose of enhancing non-proliferation introduces uncertainties that affect the performance of the safeguards system. The processes within a State that Integrated Safeguards seeks to monitor are not clearly defined. Neither are the measurement and evaluation systems for the verification of the absence of undeclared activities. A refinement of the existing diversion process model is needed that takes into account the likelihood that a State would undertake a clandestine program.

To develop such a model this paper examines the nuclear programs of seven States that have built or attempted to build nuclear weapons and one that is being accused of having one but denies it. Analysis of the factors that are essential in the decision of a State to embark on a nuclear weapons program indicates leads to the identification of certain indicators for clandestine nuclear weapons programs. It identifies the conditions and interdependencies that most likely lead those States to undertake clandestine weapons programs and develops a set of likelihood indicators that can be used in the application of safeguards to a State as a whole. Existence of serious conflicts involving territorial disputes, transparency in the social system, militarization of the State, and democratization of the political process are the common characteristics of those States. Although these are not easily quantifiable indicators, concepts such as fuzzy logic can be used to construct likelihood estimators for the presence of absence of clandestine programs leading to a State-specific safeguards approach adapted to the conditions of each State.Thus, improving the effectiveness and efficiency requires a paradigm shift to adaptive safeguards.

Keywords: adaptive safeguards; effectiveness; interdependencies; proliferation indicators

1. Introduction

The Model Additional Protocol expands the scope of the nuclear safeguards system to cover activities and facilities that are not part of nuclear fuel cycle, but may have some relation to it. The aim of the expanded scope is to enhance nuclear non-proliferation by strengthening the effectiveness and proving the efficiency of the safeguards system. While the purpose of traditional safeguards has been to verify the accuracy of the declarations submitted by the States, the purpose of Integrated Safeguards is verification of completeness [1], [2]. Traditional safeguards consists of monitoring specific locations at specified times although inspections could take place as deemed necessary under the concept of special inspections. In other words, the domain of traditional safeguards is well defined as shown in Figure 1. Integrated Safeguards aims to apply safeguards at the state level. In doing so, the intent is to assess the proliferation potential of each State on the basis of the additional information collected by the IAEA.

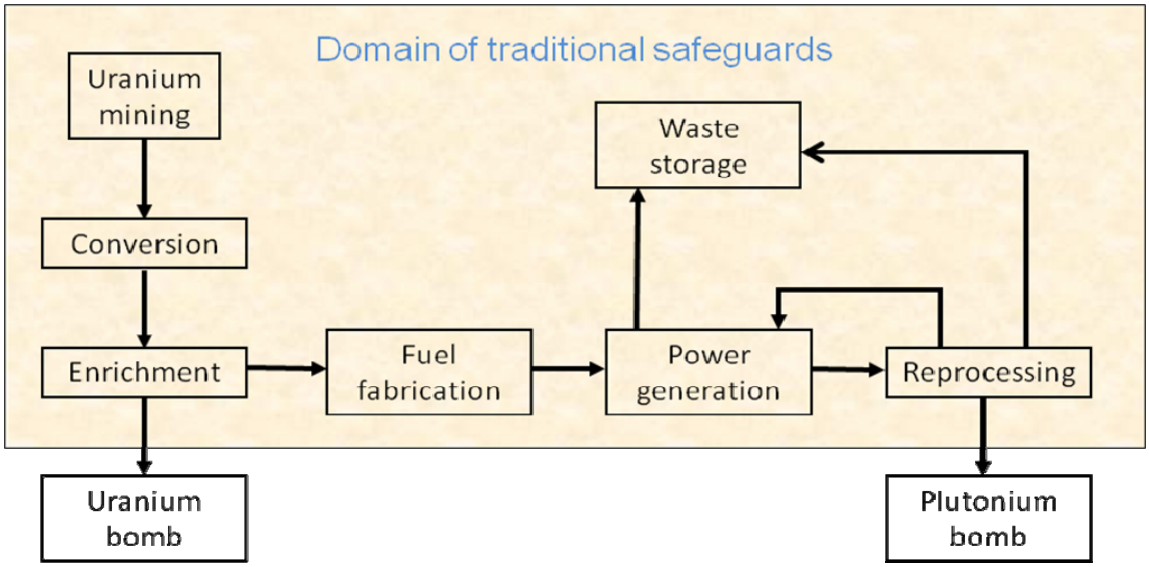

Figure 1. The domain of traditional safeguards

Looking beyond the details of the two safeguards systems, one can identify a fundamental conceptual difference between them. While the objective of traditional safeguards is clearly the detection of diversion from declared fuel cycles, the purpose of the expanded safeguards is to detect nuclear materials and activities, in other words, nuclear weapons programs in whatever form and stage of development those might be [1]. Since weapons development programs comprise a broad range of activities that can be dispersed in many locations, the quantity and types of information necessary for the application of Integrated Safeguards are much greater than those for traditional safeguards that are applied to the well defined process of the nuclear fuel cycle.

The crucial element for the good performance is the detailed definition of the observed process and the monitoring system that has been designed to observe the entire process. In contrast, under the concept of Integrated Safeguards, there is no definition of the process integrated safeguards are supposed to observe other than to verify the absence of undeclared activities anywhere in the State [3]. The absence of a model for nuclear weapons development at the state level introduces many uncertainties that affect the performance of Integrated Safeguards. As shown in Figure 2, the domain of Integrated Safeguards, in addition to the well defined nuclear fuel cycle, includes other activities such as planning, theoretical research, e. t. c., which are not as clearly defined.

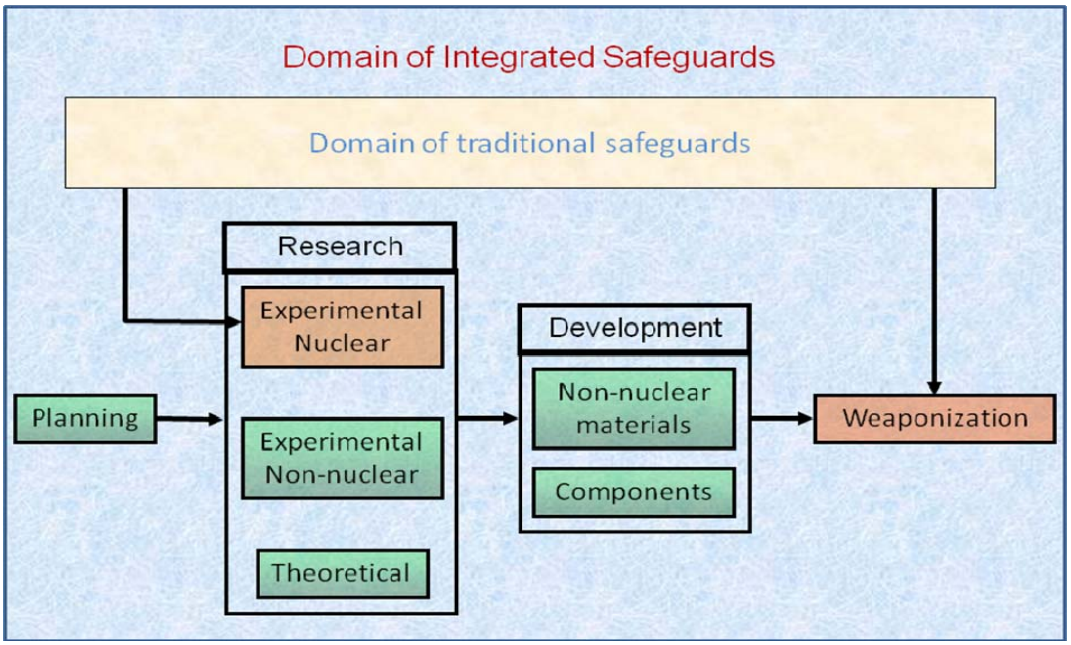

Figure 2. The domain of Integrated Safeguards includes some ill-defined elements

An important factor that is bound to play a significant inhibiting role in arriving at credible conclusions about the absence of nuclear weapons programs is the qualitative nature of some of the information acquired under the terms of the Additional Protocol. One approach to overcome this difficulty utilizes fuzzy logic concepts [4], [5]. Another uses of probabilistic models [6], [7]. Both aim to identify critical elements in various stages of the fuel cycle that could be used as indicators of diversion. The presence of one or more such indicators would be sufficient to detect violation without the need to do a complete materials balance. These techniques are potentially useful when they are applied to well-defined processes such a reprocessing plant or similar facility. It is not clear how they would apply to a distributed nuclear weapons program at the early stages of its development where nuclear materials are not involved, but all the components of the weapon are designed, assembled and tested.

An alternative approach toward the development of procedures for implementing Integrated Safeguards is to develop a model of nuclear weapon proliferation. The existence of one or more models of probable proliferating States would aid the IAEA in optimizing the allocation of its resources effectively and efficiently. The approach presented in this paper is to develop profiles of proliferating States using empirical data.

2. The proliferation landscape-States with nuclear weapons programs

To develop an effective detection system for nuclear-weapon proliferation a fairly accurate model of the process is needed. Using as a reference point the desire to prevent "wider dissemination of nuclear weapons" as articulated in the preamble of the NPT, a profile can be constructed of the state of proliferation from the time of the signing of the NPT to the present. This time line spans more than forty years of experience, an interval sufficiently long for providing enough data to form a reasonably good snapshot of the proliferation process. A State that has undertaken development of nuclear weapons is a proliferating State regardless whether or not it has signed and ratified the NPT. Following is a brief history of nuclear weapons programs of States, excluding the five Nuclear Weapon States, that either have acknowledged the existence of such programs, have been found to have clandestine nuclear weapons programs. It could be argued that the Nuclear Weapons States should also be included in the set of States used in the development of such a profile. For the purposes of this paper they have been excluded, because the concept of nuclear proliferation, by definition, uses these States as a reference point. It would be appropriate to include them for developing a model for nuclear weapon States in general.

2.1. The South America landscape

Argentina started a nuclear research program in 1953 [8], [9]. It is not clear whether the purpose of the program from the beginning was to develop nuclear weapons, or that the program started as a nuclear energy research program and subsequently branched into a secret weapons program. In the late 1960s Argentina had built a small scale reprocessing facility to produce plutonium and in 1978 had constructed a secret facility for the production of highly enriched uranium. The program was formally terminated in 1983 when a democratically elected government replaced the military junta that had taken control of the country since 1978. The impetus for starting such a program was provided by the concern that Brazil might be having such as program. In other words, the motivation was for Argentina to develop nuclear weapons as a deterrent against a perceived nuclear threat from Brazil. The concern was put to rest when democratically elected governments in Argentina and Brazil agreed to cooperate on peaceful uses of nuclear energy. Argentina terminated its weapons program, because the need to have such weapons vanished. Argentina became a party to the NPT in 1995, considerably later after it stopped its nuclear weapons program.

The nuclear weapons program in Brazil proceeded in parallel with that of Argentina [8]. Brazil had started conducting nuclear research as early as 1935 [10]. By 1953 its nuclear research program had both civilian and military components, with the various branches of the military developing their own secret programs. As in the case of Argentina, rivalry between the two neighbours was the primary motivation for Brazil's pursuit of nuclear weapons. As relations with Argentina improved, in 1990, Brazil renounced its interest in developing nuclear weapons and acceded to the Treaty of Tlatelolco in 1993 and to the NPT in 1998. Brazil accepted IAEA safeguards in its civilian facilities in 1997, much later than the date when it formally terminated its nuclear weapons programs.

2.2. The South Asia landscape

The beginning of India's nuclear weapons program may be traced to its beginning as an independent State after the colonial rule. In 1944, Nehru declared that "if India is threatened she will inevitably defend itself by all means in its disposal" [11]. One can surmise that India perceives the threats to emanate from China and Pakistan due to the unresolved border disputes. As early as 1958 India started to work on plutonium reprocessing and intensified its effort to develop nuclear weapons particularly after the first Chinese nuclear test in 1964 [11]. The first nuclear weapons test was done in 1974 followed by more tests in 1998. In total India has conducted six nuclear tests, but the size of its nuclear arsenal is not known.

It is easy to see the causal relationship between the nuclear weapons program of Pakistan and that of India. Like India, Pakistan has not signed the NPT. Nevertheless, after India had made substantial progress in developing nuclear-weapons capability, Pakistan embarked on a similar program in the early 70s following its defeat in the war with India in 1971. More so than India, Pakistan's nuclear program has benefited from information received from sources outside Pakistan. China has an ongoing cooperation with Pakistan in the nuclear field. Even more important is the reprocessing know-how A. Q. Khan obtained surreptitiously from URENCO and took it with him as he went to Pakistan to lead the fuel enrichment program. In 1998 shortly after the Indian nuclear tests, Pakistan also conducted its own nuclear tests. For the tests to be conducted shortly after those of India, it would be reasonable to conclude that Pakistan had already built all the components and made the final assembly in reaction to the tests by India.

2.3. The Middle East landscape

The nuclear landscape in the Middle East is directly affected by the unresolved Palestinian issue and the border dispute between Iran and Iraq. The three major actors are Israel, Iran and Iraq, although other States may have also considered the option of nuclear weapons.

Israel has not signed the NPT and has not formally acknowledged that it has nuclear weapons, although there have been indicators pointing to their existence. Nuclear research in Israel started as early as 1949 [12]. In 1986 Mordechai Vanunu, a technician at the Negev Nuclear Research Center, revealed that Israel had built and possessed nuclear weapons. There are suspicions that the flash detected by the VELA satellite over the Indian Ocean in 1979 was a nuclear test conducted by Israel, possibly with the cooperation of South Africa. The suspicion is that Israel's nuclear weapons program started a short time after it had signed the first nuclear cooperation agreement with the United States in 1958.

The discovery of a secret nuclear weapons program in Iraq provided the impetus for the re-evaluation of the original safeguards that were applied to declared facilities. The discovery was the more jolting by the fact that the clandestine program was taking place in a facility that was under IAEA safeguards precipitating a re-examination of the effectiveness of the facility-oriented approach. Iraq signed the NPT in 1968 and ratified it in 1969. The IAEA started applying safeguards in 1972. Shortly after the bombing of the Osirak research reactor by Israel in 1981, Iraq embarked on a crash research program to develop nuclear weapons capability and started a uranium enrichment program using indigenous technology [13]. In spite of the IAEA inspections the program was not detected for two main reasons. Some technology was not imported, but developed indigenously using commercially available components, while other was purchased in the black market; also, indigenous technical expertise was augmented by expertise procured in the international market. After the defeat of Iraq in the Gulf war of 1991, the IAEA found Iraq in violation of its safeguards obligations, because it had carried out research and development enrichment and reprocessing activities both at declared and undeclared facilities [14]. UNSCOM began wide-ranging inspections that discovered the extensive weapons development program. As in the case of Israel, the details of Iraq's nuclear weapons program were revealed in 1995 by an insider, a defecting official of the Iraqi government. The information provided in conjunction with the intensive inspection effort on the ground allowed the IAEA to conclude that by 1998 Iraq had no longer a nuclear weapons development program. When UNMOVIC suspended inspections in 2003 the IAEA was again able to report that there was no evidence the weapons program had been re-started [15].
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Iran, at various times starting in the decade of the 1980s, has been accused that it is developing nuclear weapons in secret. In contrast to Iraq that acknowledged the existence of the secret program, albeit under unusual conditions, Iran has always asserted that its nuclear program is solely for peaceful purposes and that it is in compliance with its safeguards obligations. The nuclear program started in the 1970s under the reign of the Shah with nuclear research activities. It continued after the revolution in 1979. The seize of the US Embassy in Tehran that year precipitated a confrontation with the US that lasts to this date. The first sanctions were imposed after the seizure of the Embassy followed by another set after the bombing of the U.S. Embassy and marine barracks in Beirut in 1984 and the labeling of Iran as "sponsor of international terrorism". Additional US sanctions were imposed in 1992, 1995, 1996 and 2000 amid accusations that Iran embarked on a program of nuclear weapons development [16]. In 1980, Iraq invaded Iran starting a war that lasted until 1988 that saw the use of chemical weapons first by Iraq and eventually by Iran. Toward the end of the decade of the 1980s and the beginning of the decade of the 1990s, Iran intensified its nuclear program that included research and development activities in reprocessing and enrichment. It sought to import technology from various sources including China and Russia and Pakistan [8]. The scope of the Iranian nuclear program became multifaceted ranging from power generation to research reactors, enrichment and reprocessing. As the import of technology became problematic due to the broadening of the sanctions regime Iran has intensified its efforts to develop indigenous capabilities for all stages of the fuel cycle. The IAEA inspections of the declared facilities since the signing of the safeguards agreement have not found any evidence of non-compliance. As late as 2009, the IAEA continued to verify non-diversion of nuclear material. However, in 2002, an Iranian resistance group revealed that Iran was engaged in nuclear activities at undeclared sites (Natanz and Arak) for which subsequently Iran submitted declarations. The IAEA also raised questions about previously undeclared nuclear material and asked Iran to suspend enrichment as a confidence-building measure. Iran did so and signed but has not yet ratified the Additional Protocol. In 2006 the UNSC passed a resolution demanding that Iran suspend its enrichment activities. Following Iran's refusal to abide, the UNSC imposed a series of sanctions that are in effect to this date [16]. In the meantime, the US has woven a web of sanctions that cover, in addition to nuclear-fuel cycle technologies, potential dual-use technologies and, practically, all transactions involving Iranian financial institutions. Following the imposition of these additional sanctions, Iran resumed and expanded the enrichment activities at Natanz. In a further escalation of the pressure on Iran there have been multiple calls in the US and Israel to bomb the Iranian nuclear facilities.

2.4. The East Asia landscape

The Korean War ended with an armistice in 1953 that divided the Korean peninsula into a north and a south part, the Democratic People's Republic of Korea (DPRK) and the Republic of Korea (South Korea); the state of war persists to this day. Since 1953 the United States has a Mutual Defense treaty with South Korea that allows, *inter alia*, the stationing of US forces in the territory of South Korea. A similar treaty was signed with Japan in 1954. The DPRK ratified the NPT in 1985 and signed a safeguards agreement with the IAEA in 1992. Initial inspections by the IAEA of the research facilities at the Yongbyong complex revealed inconsistencies with the data provided by the DPRK. As a result, the IAEA, in 1993, requested a special inspection to resolve the inconsistencies. The DPRK refused the special inspection and announced its withdrawal from the NPT. The IAEA considered the withdrawal illegal and found the DPRK in non-compliance with its safeguards agreement. It founded it again in non-compliance in 1994, after it began removing fuel from the reactor without the presence of IAEA inspectors. In response, the DPRK withdrew from the IAEA. These events led to the start of negotiations between DPRK and the United States that led to an agreement that provided for the DPRK to freeze the activities that had raised proliferation concerns and accept IAEA safeguards, in exchange for being provided with light water reactors and other energy supplies by the Korean Peninsula Development Corporation (KEDO) [17]. The agreement also provided for measures to be taken to reduce tensions in the Korean peninsula. A significant factor contributing to the high level of tensions is the heavy military presence across both sides of the demilitarized zone. In 2004 the President of the United States declared that the DPRK was a member of the "axis of evil". DPRK responded by revealing the existence of a program to enrich uranium for nuclear weapons, KEDO suspended the supply of heavy oil and DPRK expelled the IAEA inspectors. In 2003 it withdrew from the NPT and in 2006 it announced that it would conduct a nuclear test and did so [8].

2.5. The Africa landscape

South Africa had a nuclear research program long before it joined the NPT in 1991 and accepted IAEA safeguards. It had, however, been a member of the IAEA since 1957. With an extensive mining industry South Africa explored the feasibility and produced a plan for developing nuclear devices for peaceful uses [18]. In 1974, in the context of the Cold Ward and the upheavals in central Africa, the government of South Africa embarked on a secret program to develop nuclear weapons. Indications of the existence of the program were first given by the Soviet Union in 1977 when it detected preparations for underground testing through satellite surveillance. In 1979 the United States also confirmed the existence of a nuclear weapons program. Although sanctions had been imposed since 1979 they did not stop the program. South Africa abandoned nuclear weapons in 1989 as the Soviet Union collapsed and the political landscape on South Africa's north was changing. At the same time the minority apartheid regime realized that its rule would eventually be coming to an end. In 1993 the South African government announced that it had destroyed six nuclear weapons that it had built in the past. A democratic government replaced the apartheid regime in 1994.

Libya revealed almost explicitly in 1996 its efforts to acquire nuclear weapons when it cited the need for the development of nuclear weapons by the Arab States as a deterrent to a nuclear-armed Israel. While South Africa made nuclear weapons relying primarily on indigenous capabilities and resources, Libya, at the other end of Africa, attempted first to buy the weapons and failing that to import the technological capacity and manufacture the weapons domestically. The illicit nuclear supply network organized by A. Q. Khan played a major role by providing Libya with an enrichment plant on a turn-key basis [14]. Nevertheless, Libya was nowhere close to building a bomb after two decades of effort. Since 1992 Libya had been under UN as well as other unilateral sanctions for the downing of Flight PANAM 103 over Lockerbie, Scotland. In 2003, Libya began a series of negotiations with the United States and the United Kingdom seeking to end its international isolation. In the process, it revealed the existence of its nuclear weapons program and agreed to end it. Although Libya had ratified the NPT in 1975 and accepted IAEA safeguards in 1980, it was found in violation of its safeguards obligation in 2004 after it had revealed the existence of its secret program [14].

3. Characteristics of proliferating States

A close examination of the States that have been identified, or for which strong suspicions have been voiced, as having nuclear weapons programs reveals that they share some common characteristics.

Nuclear weapons have proliferated in regions where there have been festering and unresolved conflicts over long periods. There is a direct connection between most of these conflicts and colonialism. Although the unresolved conflict in the Korean peninsula does not have a direct connection with colonialism, it has its origins in the division of Europe and Asia into two political and military camps following the end of the Second World War.

In all cases the proliferating States felt threatened by one or more neighboring State. India first built nuclear weapons on the basis of the same rationale as that of the United States and the Soviet Union, namely, deterrence against its nuclear-armed adversary, China. This, however, caused a reaction, not so much by China, but by Pakistan that has been in conflict with India since its creation as an independent State. The fact that Pakistan conducted nuclear tests within a relatively short time following the nuclear tests by India leads to the conclusion that Pakistan had linked its nuclear weapons program to that of India.

Another example that nuclear proliferation is linked to the existence of a conflict is the Middle East. The all but acknowledged possession of nuclear weapons by Israel is its policy of maintaining military superiority in the region. Israel has sought to achieve and maintain overwhelming military superiority in the Middle East since its creation. It was not very difficult for Iraq to conclude that after the attack against the Osirak nuclear research reactor, the only way to achieve parity would be to build nuclear weapons. While Iraq was cognizant of its military inferiority vis-à-vis Israel, it must have calculated that it had military superiority vis-à-vis Iran that would allow it to achieve its objectives. When Iran wasn't as easy a target as Iraq had calculated, it used chemical weapons contrary to its obligations not to do so under the Geneva Convention to which it was a signatory.

The third link in the proliferation landscape of the Middle East is Iran. It is being accused of having a clandestine nuclear weapons program, but it insists that its extensive nuclear program is for peaceful uses. It is subject to unilateral and multilateral sanctions and it is being threatened by military attacks.

It is an open question whether or not Iran has a clandestine nuclear weapons program. If it does, the reasons for doing so would be very obvious; it would view nuclear weapons as a deterrent as the other States that have developed them have done.

The existence of conflict also forms the background for the development of nuclear weapons by the DPRK. However, unlike the other States that undertook weapons development programs in secret, the DPRK made little effort to conceal its program. No special skills were needed to detect the existence of the program when the DPRK withdrew from the NPT after the IAEA sought to clarify discrepancies in the original declaration. One can conclude that the DPRK has embarked on the nuclear weapons program as a deterrent to the overwhelming superiority of the South Korea-US alliance.

That conflict is an important predictor of proliferation is also shown by the cases of termination of nuclear weapons programs when tensions subsided. South Africa acknowledged that it had built nuclear weapons in reaction to the perceived influence of the Soviet Union in the region. With the collapse of the Soviet Union, the threat disappeared along with the need to have a deterrent based on nuclear weapons. In addition, concerns were raised within the South African government about the security of the nuclear weapons given the uncertainties associated the collapse of the apartheid regime. These two factors the government of South Africa to terminate the program and destroy the existing stockpile.

End of a different form of conflict is also associated with the termination of Libya's nuclear weapons program. Termination was the outcome of a cost-benefit analysis done by the government of Libya. After years of being subject to international sanctions and in political isolation following the downing of the airliner over Lockerbie, the quid pro quo for Libya to be re-integrated into the rest of the world was the termination of the programs for developing weapons of mass destruction. It should be pointed out, however, that Libya, unlike all the other States with on-going weapons programs, was not in conflict with its immediate neighbors. Thus, there was no evident need for nuclear deterrence which made the cost-benefit calculation easier.

Closely linked to the existence of conflicts is the form of government the proliferating States have. They all exhibit some degree of militarism. Although there are no clear-cut divisions, the majority are authoritarian in nature with the exception of India that has had a functioning democracy since independence while maintaining a strong military establishment. Israel and South Africa are special cases that share similar characteristics. In South Africa under apartheid, there was a functioning democracy within a subset of the population that was keeping the majority under control by force. Similarly, Israel is a functioning democracy for the Jewish subset of the population, while it uses force to control the Palestinians. Of the other States, Iraq under Saddam Hussein was a dictatorship as the DPRK continues to be. To a lesser degree Pakistan is also a militarist State, because the military has overthrown elected governments a number of times and still yields substantial influence over the affairs of State.

The development of nuclear weapons programs has also been influenced by the industrial profile of the proliferating States. Libya, with a rudimentary industrial and technological base tried the obvious way of attempting to purchase nuclear weapons. When that failed, it attempted to develop indigenous capability through a combination of domestic effort augmented with the clandestine importation of nuclear-related technologies. All of the proliferating States have set up their nuclear weapons programs combining domestic efforts with imported technologies and expertise. The best example of the latter is the transfer of expertise to Pakistan by Dr. Khan. The effectiveness of this path to the development of nuclear weapons has been influenced by two major factors, level of technological development and level of determination to build nuclear weapons.

South Africa was subject to trade restrictions on nuclear-related technologies. In spite of such embargo, it built its nuclear weapons in a relatively short period of time, less than ten years, because it possessed two important ingredients, raw material and scientific and technological strength. Similarly, the nuclear program of Israel started with imported assistance from France and the United States. It expanded relying on the combination of domestic scientific expertise and technological development. It remains active because the State has determined that it is in its national interest to maintain it. On the other hand, Iraq and Libya lacked technological sophistication and attempted to develop it specifically for the nuclear weapons program. After more than a decade of effort, they still were not

successful. One can only guess how long it would have taken Iraq and Libya to build a nuclear device had the former not been invaded and the latter not abandoned it.

At the start of their respective nuclear programs, India, Pakistan and the DPRK shared to a large extent the same scientific and technological profile with Libya and Iraq. They also followed a similar path, namely, a combination of imports and domestic development. The length of time it took each of the three States to develop their weapons can be associated with the resources available to each of the three. India tested a nuclear device in 1974 much earlier than either Pakistan or DPRK. While one can surmise that Pakistan had built and stored unassembled nuclear bombs much earlier than the time it conducted the first test, the evidence indicates that it devoted a substantial amount of resources, trade restrictions and embargoes notwithstanding.

While the industrial profile is the primary indicator of the ability of a State to build nuclear devices, it is secondary to the determination of a State to do so as the case of the DPRK illustrates. Taking the beginning of the decade of the nineties as the start of the nuclear program and the test in 2006 as another benchmark, it can be concluded that it took the country more than 15 years to build and test a rudimentary nuclear device. To do so it had to build an indigenous infrastructure for the entire nuclear fuel cycle. One then is led to conclude that if a State determines that it is in its national interest to build a nuclear weapon, it will do so. The length of time it will take from the moment the decision is made until a device is tested is inversely related to the scientific and technological resources available to a State. It will take much shorter if those resources exist, than if they have to be acquired *ab initio*.

4. Indicators of potential proliferators

From the preceding discussion it has become clear that nuclear proliferation has not occurred in a vacuum and is not likely to occur absent a set of conditions that would compel a State to start a covert program and be willing to bear the costs associated with the risk of detection. A State would initiate such a program only after it would determine that possession of nuclear weapons would serve a paramount national interest considering that the costs of such program would be substantial. In all cases discussed in this paper, the proliferating States sought nuclear weapons as a deterrent to threats, real or perceived, from one or more neighboring States. Thus the primary indicator of probable proliferation is the existence of serious and prolonged territorial disputes. A corollary indicator is the existence of substantial military imbalance between the potential adversaries. India started its nuclear weapons program to achieve parity with a nuclear-armed China and Pakistan reacted similarly to achieve parity with India. In the cases of Israel and South Africa, it was the perceived imbalance in the size of the conventional forces that nuclear weapons aimed to correct. In the case of Iraq a case can be made that nuclear weapons were sought either as a deterrent to a nuclear-armed Israel, or as means to gain advantage over its neighbors, or both.

The second conclusion drawn from the cases studies is proliferating States exhibit some degree of militarism. The apartheid regime in South Africa maintained power by force. Israel relies on military force to maintain the Jewish character of the State. The creation of India and Pakistan from the British colonies was accompanied by bloody conflicts that saw the rise of strong military establishments. Pakistan, in contrast to India, saw the rise of a strong and dominant military establishment. Iraq was and the DPRK still is governed by strong dictatorial regimes. Thus, another indicator of probable proliferation is the degree of authoritarian control the government of a State exercises over its population. A corollary indicator is the level of transparency in a State. The probability that a State would proliferate is inversely related to the degree of transparency in that State. In an open society it would be very difficult to maintain for long a clandestine weapons program.

Another indicator can be deduced from the length of time it would take a State to build a nuclear weapon from the moment it decided to do so. The development time is a function of the scientific, technological and economic development of a State. It took South Africa and, presumably, Israel, less time to build their weapons than it has taken the DPRK. One could only guess how long it would have taken Iraq to do it, had it not been forced to dismantle the program. Since the length of time it takes to build a nuclear weapon is inversely related to the degree of scientific, technological and economic development, the time window for detecting it is shorter for technologically advanced States that for States that are at some early stage of development. For long observation periods the safeguards system would have the opportunity to collect and evaluate more data over time. On the other hand, the shorter window of opportunity to detect a nuclear weapons program in a technologically advanced

State would make it difficult to collect enough data to detect it with some degree of confidence, raises the question whether such a program is detectable in the first place if the State decides to embark on it. On the other hand, the detection scheme for States with less advanced technological bases would have available more time and larger quantities of noisy data to detect weapons program with the same level of confidence as that for the technologically advanced States. Of course, the technological development of the States subject to safeguards forms a continuum do not fall into two distinct categories of development. Nevertheless, detection of weapons development can start with a two-detector system and evolve into an array of detectors to cover States at various levels of technological development.

Finally, the diverse stages of development raise the question what does one mean by non-compliance [19]. Now that we have compiled a set of indicators that would characterize a State that would likely initiate a nuclear weapons program, we can revisit the question of what does one mean by non-compliance from yet another perspective. There is no credible evidence to indicate that the nuclear weapons programs were detected when the programs were at the planning stage. One should seriously question the view that an effective safeguards system could be designed to detect a weapons development program that exists only on paper. A State with such a program would violate the spirit of the NPT. Nevertheless, if the safeguards system could not detect it, the IAEA would consider the State to be in compliance, because the safeguards system could not detect the non-compliance.

As a practical matter, detection of non-compliance would have to be concentrated at the acquisition phase. As it was previously mentioned, States built nuclear weapons by acquiring domestic research, development, testing and production capabilities. Traditional safeguards by seeking to detect diversion of nuclear fuel, aims to detect non-compliance at the stage of the weapon development process where the nuclear material would be enriched to weapon-grade level for insertion into an existing casing. A State in an advanced stage of technological development with an extensive research, development and manufacturing infrastructure could easily have the parts of one or more bombs assembled and tested minus the nuclear material with minimal probability of being detected. On the other hand, if a State starts from a minimal technological infrastructure on a research and development program for the sole purpose of building nuclear weapons, the import of specialized materials and technologies would be a strong indicator of a clandestine program and a safeguards system could be devised to detect potential non-compliance at a fairly early stage of the program. The same system would not be effective if it were to be applied to a technologically advanced State.

5. An adaptive model for integrated safeguards

The preceding analysis leads to the conclusion traditional and integrated safeguards are fundamentally different concepts. To verify the absence of a weapons program in the entire States one must design a system to detect the presence of specified and measurable activities. Even so, the examples of proliferation discussed in this paper illustrate the practical impossibility of doing so. Iraq's program was detected only after a military seize.

A safeguards system monitoring an ill-defined process, i.e., a "State as a whole" and lacking a precise definition of prohibited activities, i.e., unambiguous definition of non-compliance, can only generate ambiguous and imprecise results. Given the existence of these ambiguities, the challenge for the IAEA is to device a safeguards system that would minimize the uncertainty of detection of clandestine nuclear weapons program. As it has been shown, States embark on the development of nuclear weapons in the presence of certain conditions making proliferation a Markov process. Consequently, an effective detector of proliferation must take into account the likelihood of proliferation. In this paper we have identified a set of conditions that would increase the likelihood of proliferation.

This observation points to the need for an *adaptive safeguards* system. It would be conditioned upon the likelihood of the existence of a clandestine program and upon its characteristics. At one end of the scale, the likelihood would be high for regions that have the following characteristics: neighboring States are in unresolved conflicts involving territorial disputes, the political systems exhibit various degrees of authoritarianism and lack of transparency, the technological infrastructures tend to be weak. The smallest likelihood would be for those regions with the following characteristics: there are no major territorial conflicts between neighboring States, the political systems are democratic with relatively high levels of transparency, and have advanced scientific and technological infrastructures.

The relationships between the primary indicators and the likelihood of proliferation are shown in Figure 1. Since the empirical data are scarce, the functional relationships are indicative. The combination of the four indicators yields a single measure of the likelihood of the existence of a clandestine program as

$$\begin{aligned}\Pr\{prolf\} = &\Pr\{profl \,|\, cnfl\} \cdot \Pr\{cnfl\} \\ &+ \Pr\{profl \,|\, mltr\} \cdot \Pr\{mltr\} \\ &+ \Pr\{profl \,|\, transp\} \cdot \Pr\{transp\}] \\ &+ \Pr\{profl \,|\, tecndev\} \cdot \Pr\{tecndev\}]\end{aligned}$$

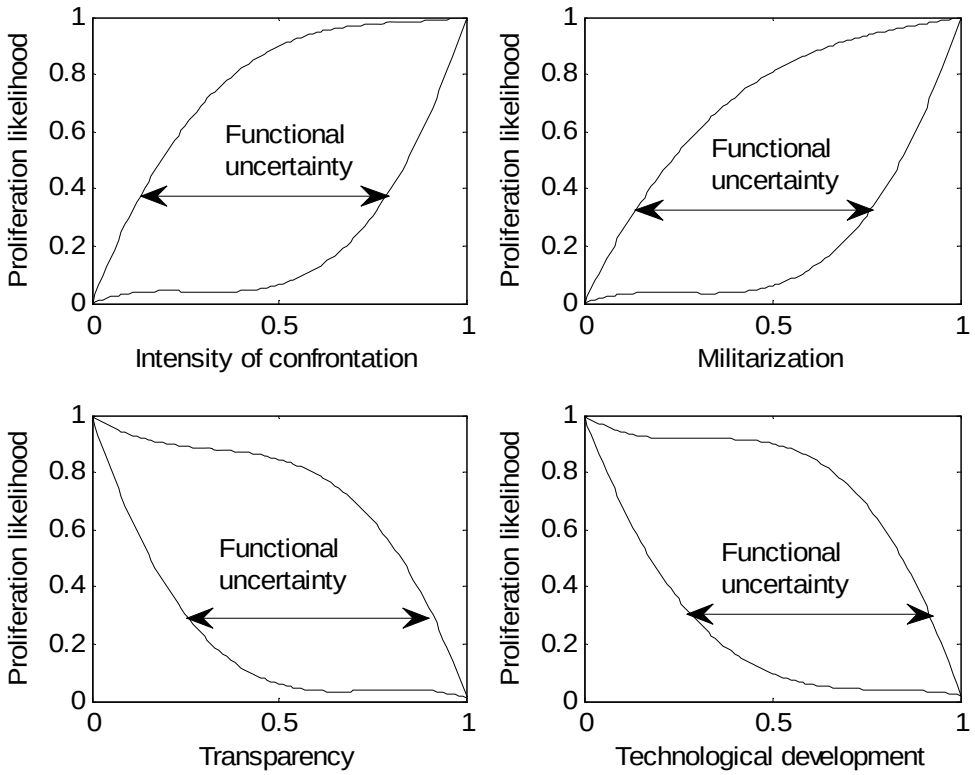

Figure 1: The likelihood of the existence of a clandestine nuclear weapons program as a function of the characteristics of a particular State

If a State is highly likely to have a clandestine program would have started it on the basis of the military significance of the weapons or on some concept of psychological warfare. Since the time it takes to develop and test a rudimentary nuclear device is more than 10-15 years, the safeguards systems has more than enough time to collect and integrate relatively weak indicators over long time intervals. As the cases of Libya and Pakistan have illustrated, the safeguards system would need to be able to monitor the imports of technology that is primarily applicable to some part of the fuel cycle. An extension of the model would be to take into account the impact of sanctions on the development of nuclear weapons. The argument in favor of sanctions is that they deprive the State suspected of developing nuclear weapons of the necessary technology and materials that are not available domestically. On the other hand, sanctions on a State determined to acquire these weapons, provide the impetus for the development of indigenous technological capabilities. All the case studies have shown that sanctions, at best, delay but do not stop nuclear proliferation. The challenge for the safeguards system is to assess the impact of restricting trade on the probability of detecting clandestine weapons programs in States that are highly likely to have such programs.

At the other end of the likelihood range, the decision to acquire nuclear weapons would be conditioned on different criteria. These are hypothetical because there has been no such case in the history of the NPT. If a State in this group decides to acquire nuclear weapons, it would have to do so if suspects that an existential threat might develop in the relatively near future for which nuclear deterrence would be a strong option. In that case, it would seek to acquire a substantial stockpile requiring the diversion of a substantial amount of nuclear material. Thus, for this group of States, the aim of the Integrated Safeguards would be to detect diversion of substantial quantities of nuclear material within a short time interval. This would be in contrast to the objective of the classical safeguards that aims to detective diversion of small quantities over long time periods. It would not be in the interest of a State belonging to that group to divert small amounts, because the material would not be of any practical use given the conditions under which the State decided to build nuclear weapons.

6. Conclusions

On the basis of the analysis presented in this paper, it can be concluded that not all States under IAEA safeguards are equally likely to become nuclear proliferators. The likelihood of a State deciding to develop nuclear weapons is conditioned on the presence of a set of identifiable conditions that would cause a State to undertake such a program. These conditions are directly related to the existence of unresolved conflicts in a region, repressive and non-transparent political systems and level of technological and economic development. For Integrated Safeguards to verify the completeness of the declaration of a State, it needs to devise a decision mechanism that would be able to detect the presence of a clandestine program using incomplete and ambiguous data. In other words a maximum likelihood estimator is needed that computes likelihood ratios on the basis of the indicators of the likely proliferators. Since conditions on the globe are not static, the detector must be flexible to accommodate the changing conditions leading to the concept of adaptive safeguards.

7. References

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The Early Years of International Safeguards – Lessons to be remembered[1]

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Abstract:

The generation of scientist and diplomats which in the sixties and early seventies of the past century took part in the creation and development of international safeguards, in partial fulfillment of the Non-Proliferation Treaty, has already left, or is leaving the scene. Therefore two of those scientists deem it worthwhile to remind the younger generation working in this field of the guiding ideas and principles which led to the safeguards system laid down in the IAEA document INFCIRC/153 in 1971. Whereas it is not our purpose to present the history of the long and controversial international negotiations which led to this document, we try to point out that the basic criteria of rationality, objectivity, formalization and transparency were the key for the universal international acceptance of the safeguards system even though it was clear already at that time that it contained serious limitations both in scope and practicality. In view of the attempts to broaden the safeguards system due to events in the nineties of the past century, it may be useful to repeat the merits of the original system and to discuss the problems which may arise when the basis of the original system is departed from.

Keywords: Non-Proliferation Treaty, Verification, Deterrence, Quantification

1. Introduction

The history of those events and international negotiations which lead to the ratification of the Treaty on the Non-Proliferation of Nuclear Weapons (NPT) and to the agreement on the IAEA safeguards system as laid down in the IAEA document INFCIRC/153 in 1971 [1] has been documented at various occasions by competent authors, see e.g. [2], [3], and it shall not be repeated here. It should be emphasized, however, that it was a unique achievement in the long history of arms control and disarmament (ACD); despite the inbuilt problems of this treaty and its verification system, some of which will be discussed subsequently, until today and more than forty years later it is still considered the most important pillar of ACD in general.

[1] The views presented here are solely those of the authors of this paper.

Instead of delving into historical detail, it is tried here to describe the reasons, i.e., the criteria and objectives which rendered this achievement possible. In fact, one should remember that at the time of the negotiation of the treaty the world was in the middle of the Cold War, and that the limitation of the number of States owning nuclear weapons implied the division of the world into States with and without nuclear weapons ("Nuclear Weapons States" NWSs, and Non-Nuclear Weapons States NNWSs). How could sovereign States accept such a division? Even though the Treaty contains a promise in the part of the NWSs to disarm, it does not contain a time line for the reduction and elimination of nuclear weapons; indeed, despite considerable efforts in that direction in the past, the world is presently still divided into NWSs and NNWSs. Worse still, the NNWSs were expected to agree that their growing peaceful nuclear industries were inspected by an international organization, whereas this was expected from the NWSs only on a voluntary basis. How was it possible to convince NNWSs to accept all this?

In the following we will point out that it was not only the good will of the NNWSs to accept these imbalances in the interest of the peace of the world, but also the rationality of founders of the safeguards system, which resulted in a wise limitation of objectives and means, and which in this way paved the road to its universal acceptance. We will try to highlight the merits of this system, and we will discuss some of the problems which may arise in departing from it.

2. Principles of International Safeguards

As a result of the long and controversial negotiations it was it was agreed that just the declared nuclear material of the peaceful nuclear industries of the NNWSs was to be subject to international inspections carried out by the International Atomic Energy Agency (IAEA) in Vienna, Austria. Furthermore, for that purpose not all areas in all nuclear installations could be inspected; instead this had to be done only at well defined key measurement points ("strategic points"). Although the possibility of special inspections was foreseen for very special occasions, this constituted a very strong limitation on the overall level of inspections, and we will come back to this issue.

Safeguarding the declared nuclear material meant establishing material balances in well-defined intervals of time (material balance periods): In each of the so-called material balance areas the initial physical inventory had to be measured at the beginning of the interval, and during the interval the inputs and outputs of material had to be measured, which together with the initial inventory formed the so-called book inventory at the end of the period. Finally, this book inventory had to be compared with the ending physical inventory, and it had to be decided whether or not a difference between those two inventories ("material unaccounted for" MUF) could be explained by measurement errors or process hold ups.

Also it was agreed that the verification of the flow of the declared nuclear material was to be performed in such a way that the operator of the nuclear plant under consideration measures the nuclear material, i.e. creates all data necessary for the establishment of the material balance, that he reports these data to the national or regional authority which then reports them to the IAEA, and that the IAEA verifies these data with the help of data generated independently and on a random sampling basis.

Finally, containment and surveillance (CS) measures were agreed upon as important complementary measures.

Before going on it should be mentioned that, originally, two approaches to safeguards were proposed and discussed, namely a game theory approach and a material balance approach. Though both of them had their merits and disadvantages, the material balance alternative was finally approved due to the aforementioned reasons of objectivity, clarity and transparency. It was also important that the material balance approach was more understandable for politicians who finally made the decision.

3. Quantification

What was the reason for this very special design of the international safeguards system to be implemented by the IAEA, and why were such important elements of proliferation as undeclared

nuclear material and undeclared nuclear facilities, technological capabilities and other aspects not taken into consideration? The answer is that it met the generally accepted criteria of rationality, objectivity, formalization and transparency which need not be explained here again in detail. On the basis of these criteria it was agreed that any statement of the IAEA at the end of a year on compliance or non-compliance of a State with the provisions of the NPT had to be based on quantitative data, not on vague and questionable impressions obtained from subjective observations of personal and activities. It was generally accepted that material accountancy and data verification using random sampling procedures provide the basis for a quantitative statement about the presence or absence of declared nuclear material at the end of the year.

Of course there exist, as already mentioned, statistical measurement errors and other sources of uncertainty which cause problems when evaluating the MUF, and there are random sampling errors especially in case of small sample sizes, but mathematical statistics in all its forms provides the tools for dealing with these quantification problems. If the distributions of the measurement errors are known, e.g. due to long term experience, and if the sampling procedures are well defined, then quantitative statements of the following form can (and have to) be made: Given an error first kind (false alarm) probability. Then a diversion of a given amount of declared nuclear material will be detected within a given time with some probability. And also, equally important in times of mutual suspicion like during the Cold War, a corresponding statement on non-diversion, i.e. compliance with the NPT, can (and has to) be made.

Two remarks are important before proceeding. First, it should be kept in mind that the measurement of nuclear material in all its forms – gaseous, liquid, solid, metallic or in compounds – is a very demanding task; indeed large and costly efforts have been made by R&D organizations throughout the world in the last thirty years and under the auspices of the IAEA to develop the appropriate chemical and physical measurement instruments and techniques both for destructive and non-destructive analyses of nuclear material. And furthermore, of equal importance was the development of a data processing and evaluation system for the IAEA which was capable to deal with the vast amount of data flowing into safeguards headquarters.

Second, when determining the probability of detecting the diversion of some given amount of nuclear material within some time, it has to be assumed that such a diversion will be performed by the operator of the facility under consideration in such a way that the probability of detection becomes as small as possible. Conversely, the inspector will adjust his inspection, e.g. his sampling plan, in such a way that this probability of detection is maximized. This means, in other words, that both, operator and inspector, behave strategically, and therefore, in addition to mathematical statistics also game theory has to be applied, see, e.g. [4], since this theory quite generally deals with these kinds of strategical problems[2].

4. Deterrence

In the basic safeguards document INFCIRC/153 the ultimate objective of NPT verification is stated as follows: …deterring by the risk of early detection…. . Before discussing this statement in quantitative terms, two other key concepts shall be introduced which occur again and again in safeguards documents and discussions, and which are closely linked to deterrence, namely efficiency and effectiveness. In words we say that a safeguards system is effective if it fulfills the objective given above, and it is efficient, if it achieves this with as low costs - manpower and finances– as possible[3]. But how do we relate this to our quantitative statements explained before?

In quantitative terms we say that the safeguards system (or some component of it in terms of facility and State) is efficient if for a given error first kind probability and a given detection time some detection

[2] If we assume that the operator looks for that strategy which minimizes the probability of detection, and the inspector looks for that strategy which maximizes it, we consider a zero sum game with the probability of detection as payoff to the inspector. A solution of this game is called a saddle point which means a pair of strategies with the property given above.

[3] The IAEA Safeguards Glossary [5] uses these terms without defining them. We follow Webster´s Online Dictionary which states inter alia: Being *effective* means to be able to accomplish a purpose. Being *efficient* means being effective without wasting time or effort or expense.

probability is achieved with as low costs as possible. As regards to effectiveness we have to quantify the term risk mentioned above. We define risk as expected gain or loss, and consequently we define that a State is deterred from illegal behavior if his expected gain in case of illegal behavior is smaller than in case of legal behavior[4]. It should be mentioned, however, that because of the requirement that no State shall be discriminated, possible gains and losses of States in case of legal and illegal behavior have never been evaluated, since this would have led immediately to some kind of discrimination.

Thus, it should be held in mind that in the framework of INFCIRC/153 efficiency is well defined and has extensively been evaluated in the past. Deterrence and effectiveness are well defined in this framework, at least conceptionally, as well, but they have never been operationalized due to the reasons mentioned above.

5. New Developments

Due to events in Iraq, North Korea and other States in the early nineties of the last century, an initiative was undertaken by the IAEA in order to broaden the objectives and tools of IAEA safeguards in order to meet the challenges posed by these events. It was argued that the subject of IAEA safeguards can no longer be just the declared nuclear material in declared facilities, and that the traditional tools of safeguards should be complemented by other means. The results of this initiative were laid down in the so-called Additional Protocol published in 1996 as IAEA document INFCIRC/540 [6].

There are many examples in many fields of science and technology where, even though there were good reasons for modification, the extension of some well defined system raised problems which did not exist in the former system[5]. In the case of IAEA safeguards the problems are obvious, let us just mention some of them.

Environmental sampling is a new technique for detecting undeclared illegal nuclear activities. How can one quantify its use for IAEA safeguards? More precisely, how many samples have to be taken and analyzed in order to reach some probability of detecting some undeclared activity in any place of the world? The question “How much is enough?” was posed already many years ago by ACD experts [8].

The control of so-called sensitive technologies is even more difficult. Here, it is already a problem to define what sensitive technologies are, and whether or not their trade serves illegal purposes in the sense of proliferation or other, peaceful ones. However if one can no longer quantify the value of some measure, then its efficiency can likewise no longer be determined with respect to some objective, and then the requirement of rationality is no longer fulfilled.

Another illustration: Once in a discussion on these issues it was stated “You will admit that more information is better than less information for international safeguards!” This is, however not true in general, and there are convincing examples which demonstrate this. Take, e.g., a material balance period of, say, one year in length. Then at first sight it seems plausible that intermediate physical inventories (more information) would improve the overall probability to detect the diversion of nuclear

[4] If we introduce gains and losses of the operator and consequently also of the inspector, then we have to consider non-zero sum games. The solution concept of these games is the so-called Nash equilibrium - a generalization of the saddle point - which is defined as a pair of strategies with the property that any unilateral deviation from that pair does not improve the deviator´s payoff [7]. Using these concepts we may say that a safeguards system is effective, i.e. the State is deterred from illegal behavior if his equilibrium strategy is legal behavior.

[5] Nuclear energy itself is an example: When it was introduced in large scale in order to meet the needs of growing electricity consumption, problems (safety, final storage of fission products) surfaced which did not exist when only fossil fuels were used. Quite a different example is game theory which was mentioned here already several times: The consideration of non-zero sum games requires a generalization of the solution concept of zero-sum games. The Nash equilibrium, which represents this generalization, is however not unique in general and poses problems of equilibrium selection which do not exist in this form for zero sum games.

material during that period. In fact it can be proven that these additional physical inventories lead to a smaller detection probability compared to that obtainable without them [4].

Thus, it is not only questionable to introduce new measures, the usefulness of which one cannot quantify and thus rendering the system less transparent, but, as the example given above shows, by using such new measures one may even reduce the efficiency of the system.

Seen from the standpoint of the NNWSs party to the NPT, the new measures raise some serious problems: Whereas their value when applied to so-called rogue States cannot presently be quantified, this very same ambiguity places well-meaning States under continuing suspicion[6]. The consequence may be a loss of confidence in safeguards measures and even a loss of interest in cooperating with the IAEA.

6. Summary

The IAEA is a scientific-technical institution with an undisputed and internationally recognized competence in all areas of the peaceful uses of nuclear energy. When, in the sixties of the last century, it accepted the extremely challenging task to organize international safeguards in partial fulfillment of the NPT, it planned and organized the work in a way consistent with its competence and its mission. This led to a safeguards system which was universally accepted even though it was always clear that it contained limitations which could not be overcome by means and tools available to the IAEA.

Due to events in the early nineties of the last century it turned out that these limitations of the IAEA safeguards system became a problem to international security. Therefore it was mandatory that attempts were made to overcome these limitations, and indeed enormous efforts, carried by the good will of those who participated, have been spent for that purpose. In fact, it may turn out one day that results of these efforts become useful for NPT safeguards or other control regimes.

Far from wishing to criticize these efforts in general, and without being able to provide concrete proposals for the solution of the problems just mentioned, which meet the objectivity criteria presented before, let us finish by repeating here our message in short.

There were good reasons for the design of the original IAEA safeguards system, and the limitations of that system were well-known. Also there is general agreement that the material balance approach and the IAEA accounting activities continue to be the backbone of IAEA Safeguards. Therefore, sufficient attention should be devoted to it, and the necessary resources in financial, technical and human terms should be deployed to keep IAEA safeguards accounting at the highest modern standards.

Indeed, later it became necessary to try to overcome these limitations. However if for that purpose one introduces elements into the original IAEA safeguards system which neither meet the high scientific-technical standards of the IAEA, nor satisfy some of the generally accepted criteria of rationality, objectivity, formalization and transparency, it may happen that they do not really help to overcome the limitations of the original system. On the contrary, they may be counterproductive in view of safeguards objectives. In fact, these elements may damage the reputation of the original safeguards system and thus, even that of the IAEA.

Even worse, and we sincerely hope that all this will not happen, well-meaning and cooperative NWWSs may loose their interest in a safeguards system which puts them under continuing suspicion. Since, as mentioned, the NPT and its verification system is considered one of the pillars of ACD, this may damage on the long run the general idea of ACD itself.

7. Acknowledgements

[6] This problem is well known in any court trial, where a balance has to be found between confirming the innocent and punishing the criminal. If a judge jails all villagers because there is an unidentifiable criminal among them, the criminal will be punished for sure, but nobody will consider this procedure to be fair.

The authors would like to thank Morton J. Canty, Forschungszentrum Jülich, Germany, for very valuable corrections and suggestions.

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Twenty years of ABACC. Accomplishments, lessons learnt and future perspectives

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Abstract:

From the inception of the implementation of the Quadripartite Agreement (INFCIRC/435), in 1991, ABACC has had an important role at the non-proliferation agenda and has also been an active player in the international safeguards. It was necessary for ABACC to develop a technical capacity to face the challenges to be a safeguards agency and to gain credibility in the nuclear safeguards world. This capacity means to develop and implement safeguards systems in the technical area, in the inspection framework, in the conceptual analysis of processes and approaches and in the political scenario. These tasks conducted the strategic plan of ABACC on the last 20 years.

Among the accomplishments, ABACC has been involved in the application of safeguards to sensitive and complex installations, in developing safeguards instrumentation, in establishing a technical and trained inspectorate, in constructing a cooperative and coordinate environment with IAEA for safeguards application. Other challenges as R&D of equipment and quality assurance systems were also managed during all these years.

ESARDA is one forum that ABACC is involved and always shares experience and ideas. On July 18th, 2011 ABACC will formally complete 20 years. This paper summarizes the accomplishments, lessons learnt and future actions for strengthen the ABACC safeguards role. It also addresses the collaboration of ABACC with other organizations in the non-proliferation and international safeguards arena.

Keywords: ABACC, Regional Systems, Safeguards Agency, Quadripartite Agreement.

1. Introduction

Considering the nuclear development of the two countries, not having a comprehensive safeguards agreement in force and willing to demonstrate the world the peaceful use of their nuclear programs, Argentina and Brazil decided to establish a Common System of Accounting and Control of Nuclear Materials (SCCC) in 1990. This means that the norms of nuclear material control should be similar in the two countries and all nuclear materials and facilities should submit to the same safeguards requirements. The SCCC was formalized through the Bilateral Agreement (INFCIRC/395), which was signed in July 1991 and ratified by the Congresses of the two countries six months later. To apply the SCCC, the Brazilian-Argentine Agency for Accounting and Control of Nuclear Materials -ABACC- was created on July 18th, 1991.

The rules and boundary conditions for the application of the SCCC by the Secretariat of ABACC are designed in the Bilateral Agreement and in one specific document named *General Procedures of the SCCC,* which is approved by representatives from each country, who act as Members of the Commission of ABACC.

The safeguards measures foreseen in the SCCC refer to declared all nuclear material after a defined starting point, although the existence of undeclared nuclear material and facilities is not excluded (i.e.

facility misuse is considered). In the SCCC there is no provision to apply safeguards either to non-nuclear materials or to research and development activity that does not involve the processing or storage of nuclear materials. The safeguards measures establish by the SCCC complied with the comprehensive type of agreements like the INFCIRC 153.

Also in December 1991 Argentina, Brazil, ABACC and IAEA signed the Quadripartite Agreement which entered into force In March 1994. Through this comprehensive safeguards agreement the countries submit all nuclear materials in all nuclear activities to the IAEA safeguards. Under this agreement the IAEA, in carrying out its verification activities, shall make full use of the SCCC. Furthermore, the IAEA and ABACC shall draw independent conclusions, but must avoid unnecessary duplication of ABACC's accounting and control activities. For this purpose, several provisions were introduced into the Quadripartite Agreement, its Protocol, and General Part of the Subsidiary Arrangements. These provisions produced several levels of coordination between the ABACC and the IAEA and between the two organizations and the two countries. Coordination arrangements were progressively implemented during the last twenty years.

2. Legal and Political framework of ABACC

The Brazilian-Argentine Agency for Accounting and Control of Nuclear Materials (ABACC) consists of a four-member Commission appointed equally by the two countries and a Secretariat with headquarters in Rio de Janeiro, Brazil. The Secretariat consists of technical and administrative professionals appointed by the Commission, clerical staff, and inspectors. The present technical staff consists of ten people, five Brazilians and five Argentineans: one Secretary, one Deputy Secretary, two planning and evaluation officers, two operations officers, two technical support officers and two accounting officers. The higher-ranking technical officer of each country alternates annually as ABACC's Secretary. One administrative and finance manager and one institutional relation complete the team of ABACC professional staff.

The inspections are performed in a cross-national basis, with Argentine inspectors verifying facilities in Brazil and vice-versa. The inspectors do not work permanently for ABACC. They are experts who usually work for the nuclear area of countries, National Authorities, or other official organizations in each country, and are convoked by ABACC's Secretariat whenever necessary. It should be noted that the team of inspectors consists not only of people working in safeguards at a national level, but also of experts from several areas of safeguards interest (NDA, DA, design and operation of nuclear installations, etc.). Presently the inspectorate of ABACC has around 90 inspectors being half and half for each country.

Established the legal framework ABACC would not succeed without the political support from the Argentinean and Brazilian government and the international safeguards organizations, such as ESARDA, INMM, DoE, European Commission to name a few, that have supported, helped and encouraged this regional organization.

The economic resources required for the implementation of the SCCC and the functioning of ABACC was established, in a general way, by the Bilateral Agreement; both countries share the costs on an equal basis. The regular operational budget of ABACC is of some US$ 4 million per year (this figure does not include the salaries of the inspectors and consultants, which are borne directly by both countries). ABACC relies also on the nuclear structure of both countries to support its technical activities. Technical cooperation with other safeguards organizations also help to withhold the expenses on research and development of safeguards tools.

As a regional safeguards system, ABACC has also to fulfil its obligations and relationship with IAEA established at the Quadripartite Agreement.

The basic undertakings of the Quadripartite Agreement are:

- The acceptance by the Parties of safeguards on all nuclear materials in all nuclear activities, for the exclusive purpose of verifying that such material is not diverted to nuclear weapons or other explosive devices

- The IAEA shall have the right and obligation to ensure that safeguards will be applied in accordance with the terms of the Agreement.
- ABACC undertakes to cooperate with the IAEA, in accordance with the terms of the Agreement.
- The IAEA shall apply its safeguards in such a manner as to enable it to ascertain that there has been no diversion of nuclear material to any nuclear weapon or other nuclear explosive device.
- The IAEA verification shall include, inter alia, independent measurements and observations.
- The IAEA verification shall take due account of the technical effectiveness of the SCCC.
- The signatory States, ABACC and the IAEA shall avoid unnecessary duplication of safeguards activities.

This Agreement is clear with regard to the relationship between ABACC and IAEA, mentioned in the Basic Undertakings and in various other articles. Furthermore, the four Parties have signed a Protocol specifying cooperation arrangements for the application of safeguards whose principles are:

(a) the need for ABACC and the Agency each to reach its own independent conclusions;
(b) the need to coordinate as far as possible the activities of ABACC and the Agency for the optimum implementation of the Agreement, and in particular to avoid unnecessary duplication of ABACC safeguards;
(c) when performing their activities, ABACC and the Agency shall work jointly, wherever feasible, in accordance with compatible safeguards criteria of the two organizations; and
(d) The need to enable the Agency to fulfill its obligations under the Agreement, taking into account the requirement for the Agency to preserve technological secrets.

Additionally, the Protocol establishes a high level Four-Party Liaison Committee, responsible for coordinating the application of the Agreement and of the Protocol and which may appoint a sub-committee for the implementation of safeguards that should foster adequate coordination between the IAEA, ABACC and both countries.

During the past 20 years the four parties have agreed in a number of Guidelines, Joint Procedures, Cooperation procedures which casted the coordination of safeguards activities in the framework of the Quadripartite Agreement.

3. Technical capacity of ABACC (implementation)

To apply safeguards as an inspectorate, ABACC had to build a system which must have competence to fully develop, implement and evaluate safeguards measures. That means, this system must have human resources capacity, a set of well developed and useful safeguard equipment, a trained and knowledgeable inspectorate body and a good headquarters support system to integrate all data obtained from safeguards activities and to generate its evaluation.

The system must operate efficiently from the planning of safeguards measures, which will rely on the right safeguards approach, the preparation for the inspection, the instruments and equipment to be used to and the way that ABACC will treat and evaluate the data obtained.

The application of safeguards in the jurisdiction where ABACC operates has offered different challenges in a range of different technical aspects. The technical activities are mainly oriented to the verification of operator declarations of material inventories or facilities usage, as stated in the SCCC and in the Quadripartite Agreement signed by the parties involved.

The technical activities derived from the safeguards verification procedures in which ABACC have mainly worked can be categorized in five different areas:

- Safeguards Approaches;
- Inspector training;
- Non Destructive Assay (NDA);
- Destructive Assay (DA);
- Containment and Surveillance (C&S);
- Data management.

ABACC has been involved in the development of unique safeguards approaches that required from the staff significant management to support research efforts on the technologies available and on the testing of new equipment as well to develop some of them.

Since ABACC was designed to get the competence and support from the States to carry out its activities, it was necessary to increase the efforts of the countries to support ABACC's activities (for instance, they need to expand their laboratories and human resources capabilities to be able to provide ABACC with the necessary support to carry out crossed inspections in the countries). This double role of the National Authorities is not new in the safeguard's field, and contributes for the effectiveness of the safeguard system. The technical support available from the two parties embraces inspectors; consultants; equipment maintenance and calibration; preparation of standards, laboratory services and any other safeguards related study or service.

That means that ABACC has to look for and manage the necessary support from the states in order to build its system. This support shall not compromise the confidentiality of the safeguards system neither the credibility of the whole system.

ABACC has the responsibility to manage and support the necessary safeguards projects in the countries or any other institution. Even counting on the ABACC's staff composed by highly qualified and experienced technicians, ABACC does not have its own laboratories or enough technical personnel to conduct new R&D techniques for a particular application or to adapt and get acquainted with new methods and technologies to be used at the inspections.

It is also ABACC's responsibility to foster the development of the countries laboratories and expertise necessary for applying the safeguards. For instance, in order to check the status of the laboratories that analyze the samples collected by the inspectors, the ABACC technical support area keeps a permanent inter comparison program running. These comparison programs are carried out with the cooperation of other international laboratories - NBL and IAEA (Seibersdorf).

Groups of experts of Brazil and Argentina are also called by ABACC as consultants in order to discuss a particular technology whenever it is necessary. Cooperation with other institutions as DOE/USA, CEA, EURATON, JRC, IAEA, KINAC and some countries are very successful and profitable to overcome this point.

The need to have a system that could manage all data collected during the inspections and could integrate the different technical areas of ABACC conduct to the development of data based systems that will allow the management of accountancy, technical and administrative data and the production of detailed reports related to safeguards implementation.

It is important to mention that two database systems that were developed by ABACC have special importance. One is the SJAR, an accountancy system that allows the data collected during the inspections feeds the ABACC database and checks are executed and automatic reports are generated for feedback the legal flow of information. It was implemented and a Joint Use System for this software was established with the IAEA. Special files and reports are generated for feeding the IAEA database. The other system is the Operation Database System that fully integrates all planning, execution, data collected and reports from all inspection information. It's a powerful tool that is used for safeguards and technical evaluation of ABACC's safeguards system.

The technical projects are conduct together with other organizations. In fact, ABACC have the policy to develop new technological systems in cooperation with IAEA, which is an end user as ABACC, and with other organizations which have human resources or laboratories. Some of the projects are listed below:

Projects	Application	Hardware development	Collaboration Organizations
Safeguards Studies	Enrichment - centrifuge and gas diffusion	No	IAEA/DoE
	On load safeguards approach	No	IAEA
NDA	Gamma and neutron detection for visual restricted access points at enrichment cascades	Yes	IAEA/CNEN/DoE

	Gamma evaluation on hold up at diffusion plants	**YES**	**IAEA/CNEA/DoE**
	Neutron collar for unique type of fuel element with slight enrichment	**Yes**	**IAEA**
	System to verify a difficult to access spent fuel assemblies stored in a Spent Fuel Pond	**Yes**	**IAEA/DoE**
	U and Pu using bulk analysis	**No**	**CNEN/ARN/DoE**
	TMIS and SMIS analysis	**Yes**	**CNEN/ARN**
	Gamma Evaluation Codes for Plutonium and Uranium Isotope Abundance Measurements by High-Resolution Gamma Spectrometry	**No**	**EURATOM/IAEA/DoE**
	Software for Enrichment Measures	**No**	**DoE**
	Differential Peak Absorption (DPA) technique	**No**	**DoE**
DA	**UF_6 sampling methodology with alumina pellets**	**Yes**	**CNEN/ARN/DoE/IAEA**
Information System	**Load cell authentication**	**Yes**	**CNEN/IAEA**
Surveillance systems	**Secure Video Surveillance System**	**Yes**	**DoE**
	Moveable Surveillance System	**Yes**	**CNEN/IAEA**
Support Systems	**Operation database**	**No**	
	Accountancy database	**No**	**IAEA**

4. Coordination and collaboration with IAEA

In the safeguards system that succeeds from the Quadripartite Agreement, four levels exist together: the facility level, the State authority, the regional safeguards organization (the ABACC), and the international safeguards organization (the IAEA). Practically all IAEA safeguards activities in the two countries are coordinated with the ABACC or through the ABACC.

The use of Regional/States Systems for international safeguards is not new. In INFCIRC/153 type comprehensive agreements states (Art. 7) that the State shall establish and maintain a system of accounting and control of all nuclear materials, and the applied safeguards shall allow the IAEA to verify the findings of the RSAC. For this purpose, the IAEA shall perform independent measurements, and its verification shall take due account of the technical effectiveness of the State's system. Furthermore, the document foresees (Art. 31) that the IAEA, in its verification activities, shall make full use of the RSAC and shall avoid the unnecessary duplication of the RSAC/SSAC activities. These provisions should be reflected in the technical criteria adopted by the IAEA: INFCIRC/153 states that the criteria to determine the actual number, intensity, duration, timing and mode of routine inspections of any facility shall include the effectiveness of the RSAC/SSAC. In short, the clear purpose of all these provisions is to determine to the SSAC or RSAC the control of the facilities and to the IAEA the control of the SSAC or RSAC. This does not mean that the IAEA can not draw independent conclusions, but rather that, in order to draw independent conclusions, the IAEA does not need to repeat all the actions of the Regional System. Although the results from coordination of activities are in general satisfactory, a full implementation of the provisions of the Quadripartite Agreement is yet to be reached.

The Quadripartite Agreement establishes that ABACC and the IAEA shall apply nuclear safeguards in a coordinate and cooperative way. In order to obtain the maximum of efficiency and effectiveness, using the minimum effort and assuring independent conclusions from each organization, coordination and cooperation between ABACC and the IAEA while applying safeguards plays a major role.

In the last 20 years a significant effort was made by both Agencies to improve Coordination and Cooperation. Many accomplishes were reached in some areas, among which we can list:

a- Documentation framework area:
- Guidelines for inspection coordination between ABACC and IAEA; (coordination)
- Procedures for Common Use of Equipment; (cooperation)
- Procedures for special inspections; (coordination); (coordination)
- Procedures for secure communication between ABACC and IAEA; (coordination)
- Procedures for Nuclear Material Reporting from the States to the Agencies; (coordination)
- Procedures for specific inspections (sensitive installations). (coordination)

b- Concept and evaluation area:
   - Participation in special groups concerned with particular installations. Development of safeguards approach and procedures for especial installations; (cooperation)
   - Reclassification of installations by type; (coordination)
   - Implementation of new policies; (coordination)
   - Domestic transfer verification approach; (coordination)
   - Implementation of Short Notice random Inspections; (coordination)
   - Facility Attachment negotiations; (coordination)

c- Inspection implementation area:
   - Planning of Inspections; (coordination)
   - Inspections programming avoiding operational conflicts; (coordination)
   - Optimization of PDI without loosing effectiveness; (coordination)
   - The application of Joint Use of Equipment in inspections; (cooperation)
   - Procedures for specific inspections (sensitive installations). (coordination)

d- Technical and operational support area:
   - Planning of equipment acquisition between the Agencies; (cooperation)
   - Comparison of DA analysis results; (cooperation)
   - Data analysis from NDA results applied to error calculation for equipment used at facilities; (cooperation)
   - Joint training on specific inspection approaches (sensitive installations); (cooperation)
   - Joint training of inspector on equipment and procedures applied in inspections; (cooperation)
   - To use the DA results from ABACC; (cooperation)

e- Administrative Area:
   - Administrative procurement, transfer and management of samples and equipment;
   - Support to inspectors in ABACC area;
   - Payment and reimbursement of services provided at ABACC zone.

Most of the items listed above have been implemented and some of them will remain in “on going” status as long as the Agencies apply safeguards based on the Quadripartite Agreement. As long as new subject that may contribute for improving the safeguards system it comes as an active item in the coordination agenda.

5. Cooperation with European Commission and ESARDA

It is important to remark at this time when ABACC is celebrating two decades of existence the support and collaboration from the European Commission, particularly represented by Euratom - European Atomic Energy Community and the ESARDA - European Safeguards Research and Development Association.

At the very beginning ABACC looked for other organizations in which it should resemble and learn from its experience. Being the first regional system, Euratom emerged as the unique choice at the beginning. However, even though we had just one organization to function as a standard to implement the ABACC system, it was a surprise for ABACC’s staff to verify the efficiency and competence of this regional system. It was also with a great pleasure that we could find support and partnership from our older brother,

During an extensive period ABACC take advantage of the way that Euratom system was developed and the help from Luxembourg staff in how to implement ABACC’s safeguards system. Today, this spirit of collaboration remains and from time to time the two organizations get together to share their experiences. We could say that even the long distance and the slight difference on the framework could not impede an open collaboration between the two directorates,

The ESARDA also played an important hole on the tasks realized by ABACC on the last 20 years. ABACC is an observer member at ESARDA and it was allowed to participate on the ESARDA working groups since the beginning. The collaboration on the Techniques and Standards for Destructive

Analysis, Techniques and Standards for Non Destructive Analysis**,** Containment and Surveillance, Implementation of Safeguards and Training and Knowledge Management working groups were the more profitable examples of such collaboration. ABACC always participate at the main ESARDA events, like the annual meeting and the annual working groups' workshops.

ABACC is proud to recognise the support obtained and friendship developed of these the high level international safeguards institutions.

5. Lessons learnt

It is not easy to summarize the experience and lessons learnt by a regional system in it is 20 years life. Some of the features are subjective and the experience is laying on the culture of the institution. However, we may state a few points:

- The exact role of ABACC in the non-proliferation profile of the two countries. Even though the non-proliferation status is a characteristic that the state has to gain or show, ABACC function as tool for that and it has to acting always as an organization that contribute for such objective;
- The relationship among the States and ABACC and the IAEA and ABACC is unique. ABACC plays in the middle of these players and ABACC has to balance the exact hole as a regional system, understanding the peculiarities of the countries and the requirements of an international system that tends to be uniform and mechanistic;
- The IAEA needs the support of other safeguards systems to accomplish its tasks;
- The new trends on international safeguards and the new technologies should be absorbed by ABACC and after the right understanding they should be transmit to the states;
- To guarantee the credibility of the system, ABACC has to be independent of the states political line and at the same time to have it own technical criteria (that sometimes will have difference for the IAEA);
- Most important characteristic of ABACC is the technical competence and acknowledgement of this competence by the international safeguards community;

6. Future trends

ABACC considers of fundamental importance and fully supports IAEA initiatives to define the conditions and activities pursuant to the State-level concept and to define State-specific safeguards objectives to be applied to a State and to use the current Safeguards Criteria as a menu of safeguards tools, not as a prescriptive requirement for safeguards application. In particular, ABACC considers that some basic documents should be prepared for the appropriate consideration of the regional safeguards international system, in particular:

a) A complete description of the methodology for determining the State-specific Safeguards Technical Objectives that are intended to be used for a state safeguards evaluation. Such a description would allow to consider alternative safeguards activities for covering these objectives and facilitate the proper introduction of new safeguards tools, either because of new techniques (e.g. remote monitoring, environmental sampling) or because use is made of RSAC or SSAC resources.

b) A scheme of the rules and criteria to be used for evaluating the objective elements of an RSAC or SSAC. This will allow both the IAEA to consider the eventual "delegation" of some verification activities and the RSAC or SSAC to consider improvements of its system for a better inclusion into the integrated scheme.

c) A summary description of the basic rules that would be used to consider the less quantitative elements that shall be considered for evaluating the credibility and effectiveness of RSAC and SSAC. This will allow the States and the RSAC or SSAC to understand logical differences in the application of the state level concept as well as to promote changes aimed at increasing credibility and effectiveness of the local system.

d) A summary description of the basic scheme of the quality assurance program to be used by the IAEA to confirm, on a continuous basis, that the RSAC or SSAC maintains its initial credibility and

effectiveness. This will allow the States and the RSAC/SSAC to be prepared for an extensive integration. It should be noted that increase integration would imply, inter alia, the presence of IAEA inspectors at the RSAC or SSAC headquarters for long periods of time or the sudden incorporation of an IAEA Inspector to an ongoing RSAC/SSAC inspection at a given facility.

7. Final remarks

The successful projects developed by ABACC and its partners in this field of safeguards have contributed for the application of safeguards in a more effective and efficient way contributing for non-proliferation. The ABACC Secretariat is proud to complete the 20th anniversary with an organization that is recognized for excellence in international safeguards. New challenges are coming for the years ahead, and the initial spirit and strength of ABACC organization remains for facing these challenges.

8. References

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Overcoming the Substantive and Procedural Challenges of Negotiating a Fissile Material Cutoff Treaty

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Abstract

Reaching an agreement on a Fissile Material (Cutoff) Treaty (FM(C)T) remains a critical tool in the nuclear disarmament toolbox. The strategy toward that end thus far has failed. In order to address this shortcoming of the nonproliferation and disarmament regime it is necessary to rethink the players, forum, methods and alternative factors involved in making progress on such a treaty. This paper will outline a handful of approaches that could resolve the deficiencies that are allowing the current situation to be perpetuated in Geneva.

The Conference on Disarmament (CD) came to no agreement in 2010, and a FM(C)T came no closer to its actualization. The deadlock at the CD continued with little hope of waning, as it has for more than a decade. Individual country positions combined with a misuse of the rule of consensus allowed certain states to hold the proceedings hostage, despite a significant amount of political will to move forward with negotiations in the follow-up to 2009's adoption of a program of work. It is therefore pertinent to consider and advocate for the removal of negotiations from the CD to an alternative body, ideally where the rule of consensus cannot hijack the proceedings.

Even more important than issues of procedure, it is vital that the root causes of the lack of agreement between states possessing nuclear weapons (whether NPT-sanctioned or otherwise) be addressed. The main substantive issue, existing stocks of fissile materials, is touted by some members of the CD as the reason for the lack of resolution on an FM(C)T. In order to progress beyond this hurdle, the issue of the 2008 Nuclear Suppliers Group exemption to India must be considered, together with the long-term ramifications of that decision.

17 Nuclear Forensics

Nuclear Safeguards and Nuclear Forensic Analysis by Secondary Ion Mass Spectrometry

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Abstract:

A primary tool for detecting undeclared nuclear activities is sample analysis of uranium-bearing aerosol particles collected on cotton swipes.

The typical analytical task for a laboratory specialized in uranium particle analysis can be divided into two steps. The first task is a "needle in the hay stack" problem of locating sub-micron sized uranium particles in a matrix of millions of other particles. Once a particle is found, the second step follows where the isotopic composition of the particles is measured with high precision.

For many years SIMS has been used as one of the mainstay techniques for particle analysis on Safeguards samples. The SIMS instrument is unique in that it is the only technique that can both search a material to find the particles of interest and to perform the isotopic analysis within one instrument. In recent years there have been several improvements in the use of SIMS for nuclear Safeguards and forensic analysis. A major step forward has been the implementation of Large Geometry (LG) – SIMS instruments [1], and the use of the newly developed, Automated Particle Measurement (APM) image software [2]. This paper describes the recent developments in image processing used for particle identification in APM and compares the performance of different image processing algorithms.

Keywords: SIMS; LG-SIMS; uranium; particles; nuclear safeguards; nuclear forensics

1. Introduction

Secondary ion mass spectrometry (SIMS) has been a mainstay technique for more than a decade in Safeguards applications to detect undeclared nuclear activities.

Recent improvements in particle analysis for Safeguards purposes include the use of Large Geometry - SIMS (LG-SIMS) rather than the previously used Small Geometry - SIMS (SG-SIMS) instruments. LG-SIMS instruments have a larger magnetic sector radius that provides improved performance for uranium particle analysis mainly due to their high transmission at high mass resolution. Common molecular interferences that can hamper the measurement in normal SIMS analysis are removed efficiently, thus greatly improving the precision and accuracy for the uranium isotope measurements on small particles [1].

Another major improvement has been the development and implementation of the CAMECA Automated Particle Measurement (APM) software for both SG- and LG- SIMS. APM performs a fast screening over the whole sample using the ion imaging capabilities of the SIMS instrument. It provides the exact location of the uranium particles and an estimate of the enrichment for every particle [2], which is an extremely important consideration for Safeguards purposes.

Typically, swipe samples are taken at enrichment facilities to ensure that the level of enrichment remains within declared values. If a facility would start to produce higher enrichments it is essential for the Safeguards regime to have an early detection. This requires both timely analysis and the ability to find traces of more highly enriched material production in a background of material with lower enrichment. First of all, there is a "needle in the hay stack" problem since one needs to find often scarce uranium particles in a matrix of other materials. Secondly, it is essential to have an estimate of the enrichment for every uranium particle, in order to detect more highly enriched particles hidden in a background of lower enrichment particles.

The only method currently available that can effectively perform both of these two tasks is SIMS equipped with an automated image screening and processing software. An example of results obtained with this method is shown in Figs. 1 and 2. A sample with a main population of 10% enrichment was found to contain also a few natural uranium particles and at least two particles with higher enrichment. In total, three planchets were prepared and analysed resulting in the identification of 16216 uranium particles. These results were followed by more precise and accurate micro beam measurements performed on individual particles which confirmed the presence of natural uranium as well as 10%, 15% and 20% enrichments.

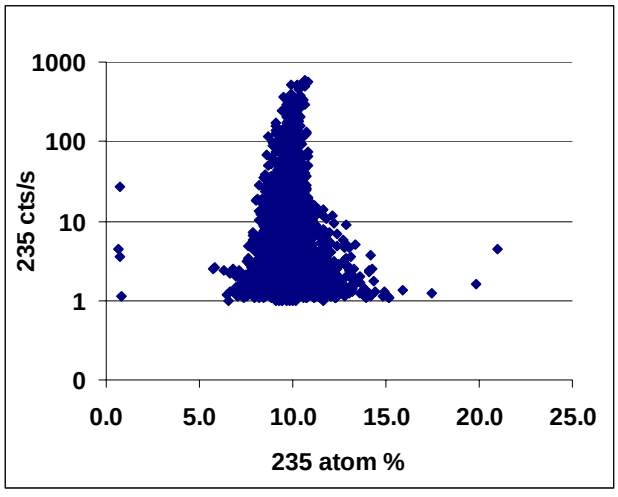

Figure 1: Scatter plot of 16216 particles, identified by APM screening measurements (data points below 1 cts/s have been filtered out).

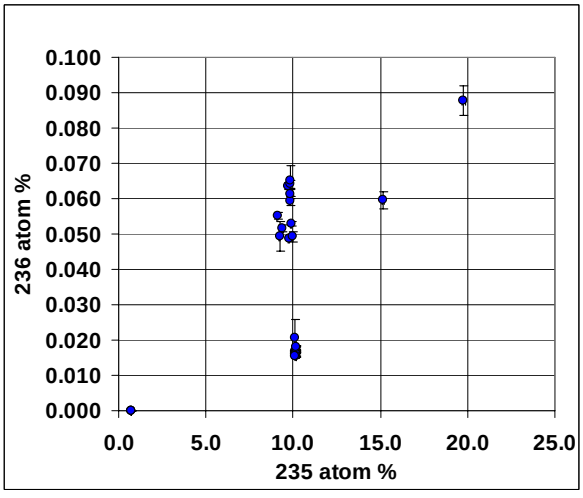

Figure 2: ^{236}U versus ^{235}U plot for 29 micro beam measurements on individual particles from the same sample presented in Figure 1. This type of measurement provides isotopic data with higher precision; results confirm the basic findings obtained from the screening measurement.

2. Description of the SIMS analytical task for uranium particle analysis

The analytical SIMS work can be separated into three main tasks:

1. The first is to remove particles from a cotton swipe and to place them with a good dispersion on a graphite planchet. The best technique for accomplishing this is by vacuuming the particles from the swipe and directly spraying them onto a planchet using a vacuum impactor. This provides a high quality deposition of the sample material [3]. The extraction efficiency from the swipe has been measured at a level of 48% for one type of impactor, and up to 67% for a second type of impactor [3, 4]. This extraction efficiency is so high that there is seldom a lack of material for the particle analysis. The main remaining issue on the sample preparation is the difficulty to regulate how much material is deposited on the graphite planchet. At present, the quality of the sample preparation mainly relies on the experience of the analyst.

2. The second task is to search through the particles deposited on the planchet to find the ones of interest. This can be done with the Automated Particle Measurement (APM) software developed by CAMECA. A previous study [2] pointed out that the main remaining issue for further improving the isotopic screening was the need for a more advanced image processing algorithm that provides more efficient particle identification. This point is discussed further below.

3. The final task is to perform accurate and precise isotope ratio measurements of the selected particles. This requires the highest possible useful ion yield during uranium measurements. SIMS has been shown to provide a high ion yield (1.0 - 1.2%) for uranium measurements using LG-SIMS with multicollector ion detector system. A current limitation of the SIMS technique is the occurrence of isobaric hydrogen interferences which represents an area of potential improvement [1].

For the highest quality analytical work, all three steps have to be optimised not only in performance but also in timeliness. Recently, the quality of screening measurements was greatly improved by implementing a more advanced image processing algorithm into the APM particle search software. This paper presents this new method and compares its performance against two different image processing algorithms.

3. Results and discussion

In order to compare the different image processing algorithms, they have been applied to an ion image obtained during a screening measurement [2]. This image is appropriately challenging due to the high density of particles in combination with a high variation of intensities (Fig. 3). Results comparing the three particle definition algorithms, using varying settings for algorithms are presented in Figs. 4 to 11,

1. **Fixed threshold image processing algorithm:** this method is implemented in the first version of the APM software. It uses a fixed threshold that must be defined by the operator; all pixels with intensity above this threshold are set to "on", and all pixels below to "off". Then, the image is scanned and any cluster of continuous "on" pixels is labelled as a unique particle. Finally, only particles with a number of pixels above the user-defined value are kept. The results obtained with this method (Figs. 4 and 5 for two different threshold values) clearly show the drawbacks of this method. First, closely situated particles are often identified as one unless the threshold is fortuitously set in the correct range. Second, if a wide range of particle sizes is present, the threshold must be set sufficiently low to include both large and small particles, which then exacerbates the first problem of particle agglomeration (Fig. 4).

2. **Niblack image processing algorithm [5]:** this method uses a variable threshold calculated at pixel (x,y) as $t(x,y) = m(x,y) + k \times sd(x,y)$ where m(x, y) and sd(x, y) are the pixel intensity mean and standard deviation values computed over a local neighbourhood of (x, y). The local neighbourhood is typically set between 16×16 pixels up to 32×32 pixels. The Niblack deviation factor k is a constant that can have values ranging from 0 to 1. A lower threshold is applied (typically at 10 counts) to remove some noise. This is followed by a smoothing algorithm that is making an average over 3×3 pixel areas before applying the Niblack algorithm. Results

obtained for two different local sizes of neighbourhood areas and different deviation factors are shown in Figs. 6 to 8.

3. **New auto-threshold image processing algorithm developed by CAMECA:** The particle identification is based on the local maxima in the image, that is, pixels that have intensities higher than all surrounding pixels. These maxima correspond to the centers of the particles. To avoid noise spikes in the image being identified as particle peaks, images are first smoothed over a block of n x n pixels (typically 3 x 3), the intensity of each pixel being replaced by the average value of the n x n block centred on that pixel. Also, background pixels, defined as all pixels with intensity below a threshold (typically 1-5% compared to maximum) are set permanently to "off" before finding local maxima. Once identified, the local maxima are sorted in descending order of intensity (I[i], i=1...N, N being the number of local maxima) and an iteration is performed for each value of I[i], starting from the absolute maximum (i=1). When starting a given iteration, particles may already be defined from the previous iterations. For a given iteration, pixels with intensity equal or above I[i] are set to "on". Then, the image is scanned in order to find the clusters of adjacent "on" pixels. This may result in an enlargement of a particle that already existed or the definition of a new particle. Then, the algorithm checks if the particle enlargement resulted in two or more particle centers being included in one unique particle. In this case, the new threshold I[i] is not applied to those particle centers, and instead threshold I[i-1] from previous iteration is kept. From that iteration on, the size of the particles associated to those particle centers is fixed. Therefore, at the end of all iterations each particle is defined with a specific threshold. Finally, only particles with a number of pixels above a user-defined value are kept. Results obtained for different noise exclusion conditions are shown in Figs. 9 to 11.

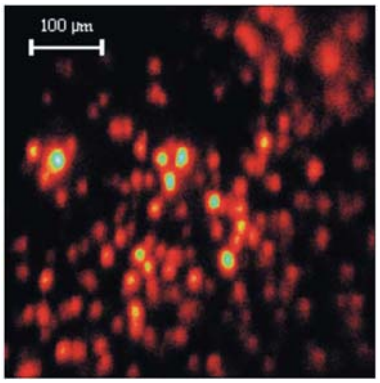

Figure 3: SIMS scanning ion image corresponding to the sum of ^{235}U and ^{238}U isotope intensities over a 500µm×500µm area. This sample is a challenge for the particle identification algorithm due to the high density of particles and the high variability in particle sizes. This image is used for the tests shown in Figs. 4 to 11.

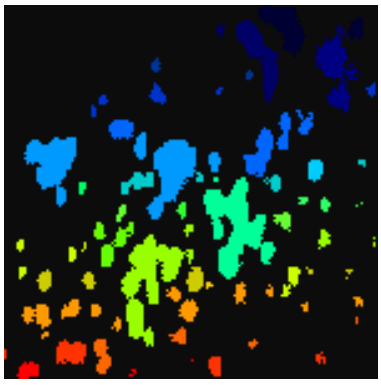

Figure 4: Fixed threshold method:

- Threshold of **50 counts**
- Minimum particle size = 10 pixels

This was the optimum setting that identified the highest number of particles (=73).

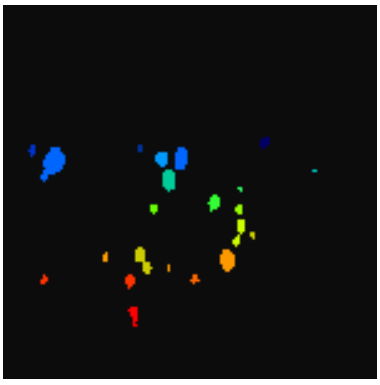

Figure 5: Fixed threshold method:

- Threshold of **200 counts**
- Minimum particle size = 10 pixels

The threshold was increased compared to previous figure in order to separate the large particles in the centre. This however resulted in only 22 identified particles.

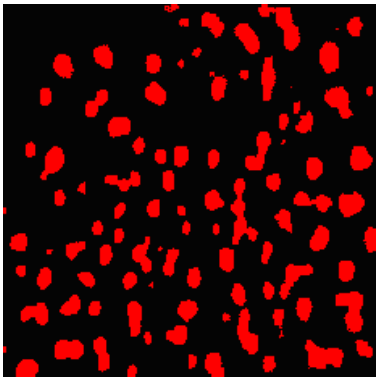

Figure 6: Niblack method:
- Deviation factor of **k=0.5**
- Threshold algorithm on **32×32 pixel areas**
- Lower threshold at 10 counts
- Smoothing average of 3×3 pixels
- Minimum particle size = 10 pixels

92 particles were identified.

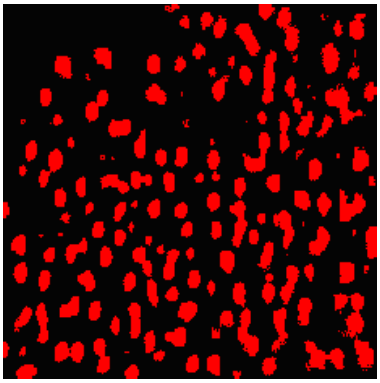

Figure 7: Niblack method:
- Deviation factor of **k=0.2**
- Threshold algorithm on **16×16 pixel areas**
- Lower threshold at 10 counts
- Smoothing average of 3×3 pixels
- Minimum particle size = 10 pixels

132 particles were identified

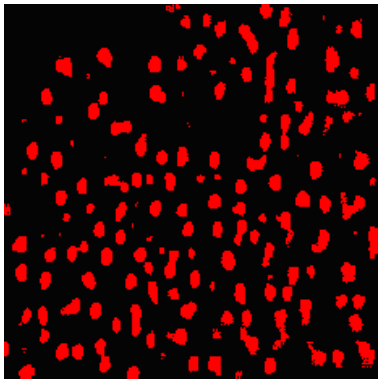

Figure 8: Niblack method:
- Deviation factor of **k=0.5**
- Threshold algorithm on **16×16 pixel areas**
- Lower threshold at 10 counts
- Smoothing average of 3×3 pixels
- Minimum particle size = 10 pixels

141 particles were identified.

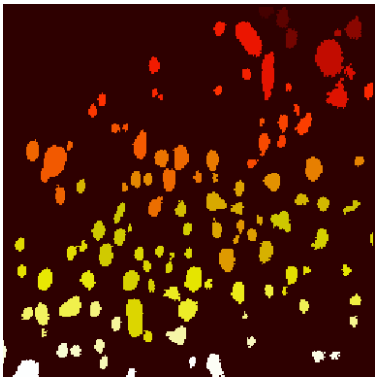

Figure 9: Auto-threshold method:
- Threshold exclusion at **5%**
- Smoothing average of 3×3 pixels
- Minimum particle size = 10 pixels

120 particles were identified.

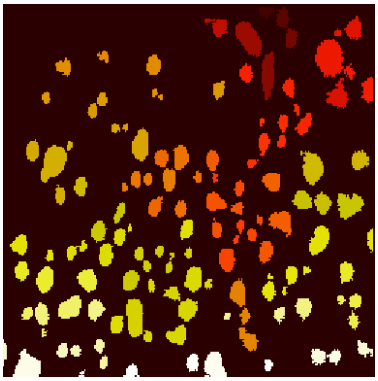

Figure 10: Auto-threshold method:
- Threshold exclusion at **3%**
- Smoothing average of 3×3 pixels
- Minimum particle size = 10 pixels

132 particles were identified.

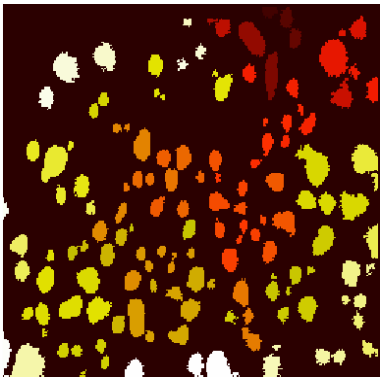

Figure 11: Auto-threshold method:
- Threshold exclusion at **1%**
- Smoothing average of 3×3 pixels
- Minimum particle size = 10 pixels

141 particles were identified.

In order to perform a comparison for particle screening measurements containing a larger number of images, a sample containing 10% (NBS-010) and 50% (NBS-050) enriched uranium particles was analyzed, see Figs. 12 to 15. All measurements were performed on a CAMECA 4FE6 IMS instrument [2].

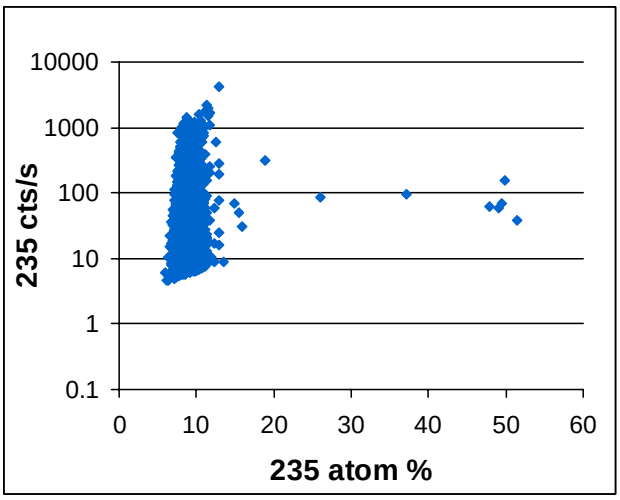

Figure 12: Results of the APM measurement on a QC sample containing 10% and 50% enriched uranium particles using the fixed threshold algorithm with the settings:

- Fixed threshold of 100 counts
- Minimum particle size = 10 pixels

The total number of identified particles was 7500. With a threshold lower than 100 counts large clusters of combined particles were obtained, whereas a higher threshold resulted in a significant loss of smaller particles.

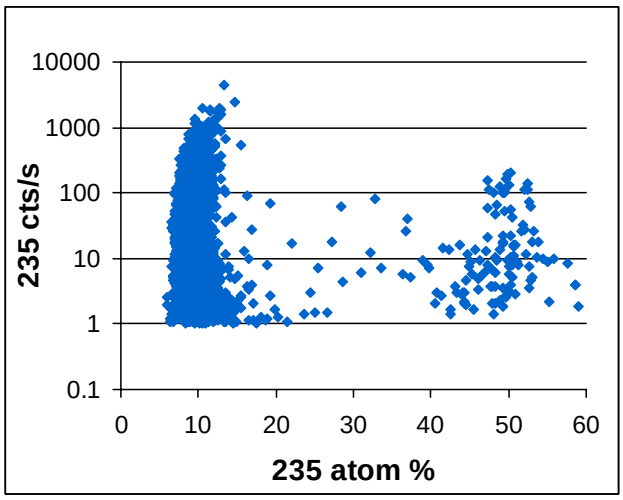

Figure 13: Results of the same APM measurement, presented in Fig, 12 but using the Niblack image processing method with the settings:

- Deviation factor of k=0.9
- Fixed threshold at 10 counts to remove minor noise
- Threshold algorithm on 32×32 pixel areas
- Minimum particle size = 10 pixels
- Smoothing average of 3×3 pixels
- Particles with ^{235}U count rates below 1 count/s have been filtered out

10691 particles were identified.

A total of 17803 particles could be found using a 16 × 16 area with a deviation factor of k= 0.5. The settings above still provided a better separation of the two enrichments.

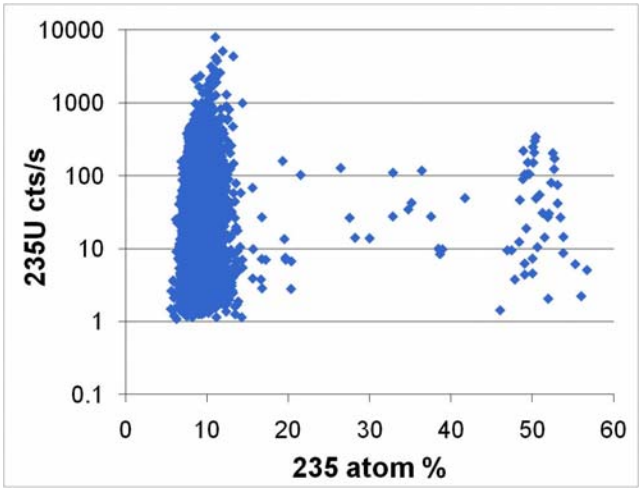

Figure 14: Results of the same APM measurement presented in Fig. 12, but using the new auto-threshold algorithm:

- Threshold exclusion at 5%
- Smoothing average of 3×3 pixels
- Minimum particle size = 10
- Particles with ^{235}U count rates below 1 count/s have been filtered out

14137 particles were identified.

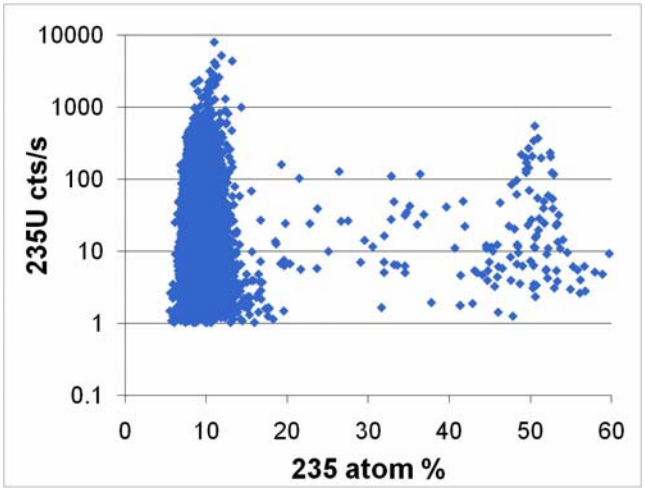

Figure 15: Results of the same APM measurement presented in Fig. 12, but using the new auto-threshold algorithm:

- Threshold exclusion at 3%
- Smoothing average of 3×3 pixels
- Minimum particle size = 10
- Particles with ^{235}U count rates below 1 count/s have been filtered out

15541 particles were identified.

From the image tests with the **fixed threshold method**, it can be seen that an increase of threshold gives a better separation of particles but results in a significant loss of smaller particles with lower count rates, see Figs. 4 and 5. When using the **Niblack method**, an increase in deviation factor k from 0.2 to 0.5 provides an improved particle separation but also results in the defined particle areas becoming smaller, see Figs. 7 and 8. A change from a 32×32 area to a 16×16 area has a significant effect in improving the particle separation, see Figs. 6 versus 8. The new **auto-threshold method** (Figs. 9 to 11) provides an improved particle definition as the exclusion level is lowered, and the threshold is optimized for each particle. This results in large particle areas, which improves the precision on the enrichment computed for each particle.

Figs. 12 to 15 compare the results obtained with the different algorithms on an APM measurement performed on a heavily loaded, mixed-enrichment sample. Results obtained with the fixed threshold show that only a few particles with higher enrichment could be detected. Instead, both Niblack and auto-threshold methods find a considerable amount of ~50% enrichment particles. In particular, results using the auto-threshold with 3% noise exclusion and Niblack algorithms provide similar enrichment distributions, with a higher number of identified particles for the auto-threshold method.

The fixed threshold method works well on most Safeguards samples that typically only contain small amounts of sub micron sized uranium particles. From the images shown in Figs. 3 to 11, it is however obvious that the Niblack and the new CAMECA auto-threshold algorithms provide significantly better results than the one using a fixed threshold in samples with high densities of particles exhibiting differing intensities. The results obtained with the Niblack method and the new auto-threshold methods are similar. The auto-threshold method, however maintains larger masks for the individual particles while still having an optimum separation, see Figs. 9 to 11. This property has the important advantage of increasing precision and thus confidence of isotope ratio estimates of individual particles. The fixed threshold and the Niblack methods have the advantage of being very fast in the image processing.

4. Conclusions

There has recently been a major advance in the development of the SIMS method for the application of isotopic analyses of uranium aerosol particles for nuclear Safeguards purposes. This leap is the result of the implementation of LG-SIMS instruments combined with the new APM software. This development has made SIMS a primary measurement technique that has unique capabilities to solve difficult nuclear Safeguards tasks in a timely manner.

The newly implemented CAMECA APM auto-threshold image processing method that has been presented in this paper represents an important improvement as it significantly enhances the particle screening performance on dense samples.

Future challenges in optimising and improving the measurement technique is likely to focus on issues like minimising the hydrogen interference and optimised calibration of the multi ion counting system detectors.

6. References

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Tracing Origins of Uranium Products

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Abstract:

In 2009 the Canadian Safeguards Support Program undertook a sampling campaign to follow a batch of UOC feed material through the refining and conversion processes at Cameco Corporation's BlindRiver and Port Hope facilities. Sixteen feed UOC samples, twenty-two samples from the refining processes, Uranium Ore Concentrate (UOC) to UO_3, and twenty-one samples from conversion processes, UO_3 to UF_6, were taken.

The samples were analyzed for trace element content by ICP-MS, uranium and lead isotope ratios by MC-ICP-MS and hydrogen and oxygen isotope ratios by IRMS techniques. The analytical results have been examined to find the extent to which trace element levels and isotopic compositions change at the various processing steps. Lead isotopic ratios, indicative of geological age, may provide information about the source of the UOC feed. Oxygen and hydrogen isotope ratios could provide further information on the source of the yellowcake as well as the reagents, conditions and methods used at each step in processing stages.

The paper will provide an update on our progress and their implications for traceability of downstream material.

Keywords: UOC; uranium; impurities; forensics; traceability; isotopic

1. Introduction

The principal objective of this work is to be able to identify the origins of Uranium Ore Concentrates (UOCs) and the down-stream uranium products leading to UF_6. Characterisation of these materials has great value for forensic work which could be required in enforcing import/export control and non-proliferation arrangements. This programwas conducted as a joint IAEA task; samples were also shared with the IAEA (Seibersdorf Analytical Laboratories, SAL) and JRC-ITU. It is anticipated that this arrangement willnot only add to their knowledge of these materials, but will also facilitate inter-comparison of laboratory results.

For a variety ofreasons, the distribution of samples and the delivery of analytical results has taken far longer than anticipated. It was only just before this meeting that complete analysis results from Canada became available. Unfortunately, we now find that different laboratories can returnquite different results for some elements. We clearly need more time to understand these differences and evaluate the available data. The findings presented here are preliminary and limited, but provide an indication of the direction of our work.

2. Collection of samples

Canada hosts uranium mines, a refinery and conversion facility. This provides the possibility of tracking the same material through from ore to UF_6, though in this case, we only tracked material from Uranium Ore Concentrate (UOC) through to UF_6.

Samples were collected at Cameco's BlindRiver facility (refinery) in November 2009 and at Cameco's Port Hope facility (conversion) in February 2010. The location of these two facilities is shown in Fig. 1.

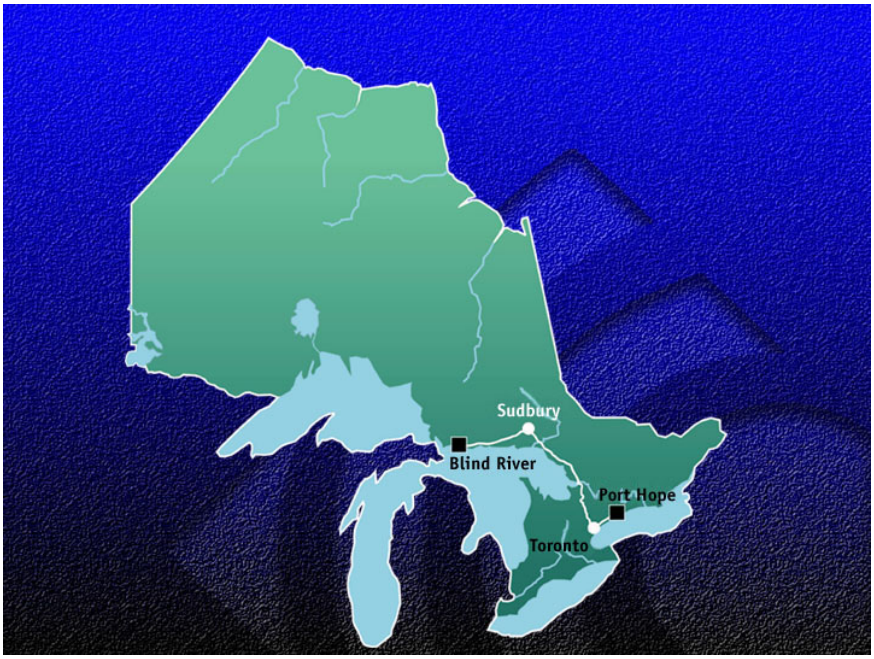


Figure 1 - Refinery and Conversion facilities in Canada. Distance by road, approx 580 kms.

Figure 2 indicates the major processes involved at these facilities. Typically, UOCs are blended prior to refining. The bracketed numbersindicatethe proportion of each UOC entering the refinery feed. Blending is necessary to ensure a consistent end product. As we will see later, the impurities present in each UOC vary considerably; blending helps to even these variations, but it also makes it difficult to track a subsequent uranium product back to a particular UOC.SixteenUOC lots were sampled,in reality only fifteen entered the production stream that was sampled.

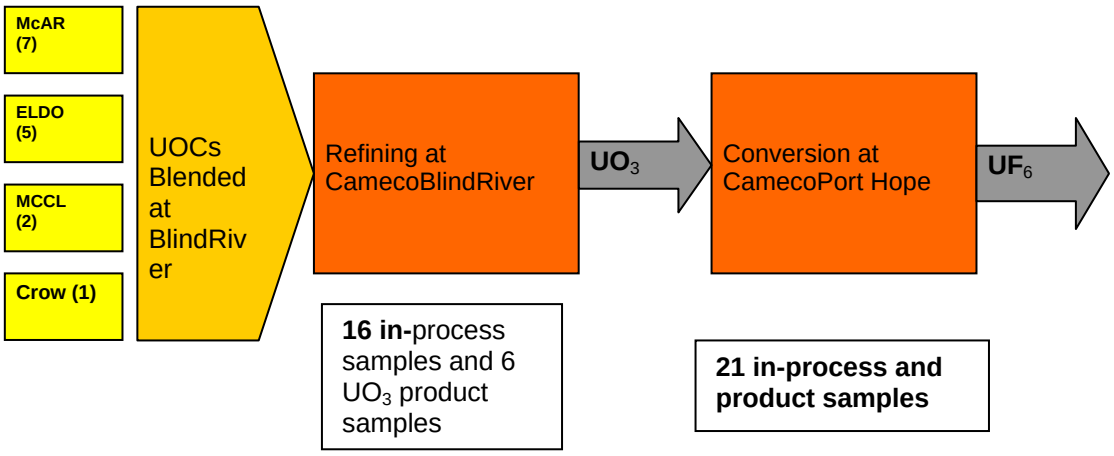


Figure 2 The major processes: blending, refining and conversion

Considerable effort was expended duringthesampling campaigns to ensure that samples would be taken from the same material as it progressed through the refinery and conversion processes.

Samples were carefully homogenised before splitting to ensure that the samples distributedto the various laboratories would be consistent.

Samples were distributed to:

- Queen's University Facility for Isotope Research (QFIR), Kingston, Canada
- Seibersdorf Analytical Laboratory (SAL), Vienna, Austria
- Institute for Transuranium Elements (ITU), Karlsruhe, Germany

.
Analysis work carried out at QFIR included ICP-MS for elemental composition and MC-ICP-MS for isotopic ratios. The isotopic standards used were UNBL129A for uranium and V-SMOW (0.1 per mil accuracy) for oxygen.

3. Initial assessment of results

3.1. Element data

An initial review of the element data available has revealed some inconsistencies. Resolving these inconsistencies will take some time; so the following results should be considered preliminary. Figures 3, 4, 5 and 6 show element impurities for the four types of UOC. The same set of elements is shown and the same vertical axis scale to aid comparison. The first is the "cleanest" for the selected elements. Figure 4 is characterised by a high sulphur content (>60,000 ppm), whereas the Figure 5 shows a broader selection of impurities. Sodium dominates Figure 6. As illustrated by other studies UOCs can be easily characterised by their elemental impurities.Figure 7 shows the level of the same set of elemental impurities for the UO_3 product; they have been reduced considerably.

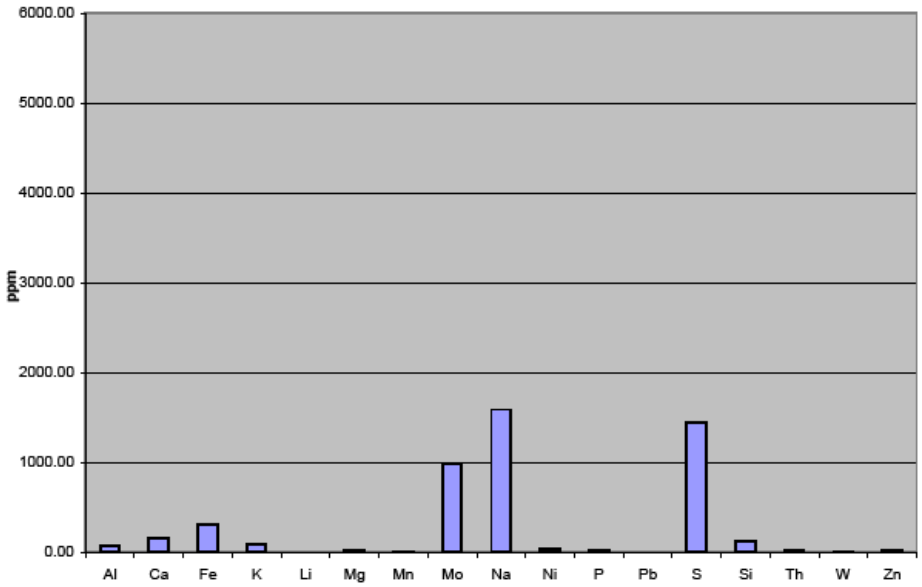


Figure 3 UOC: MCCL

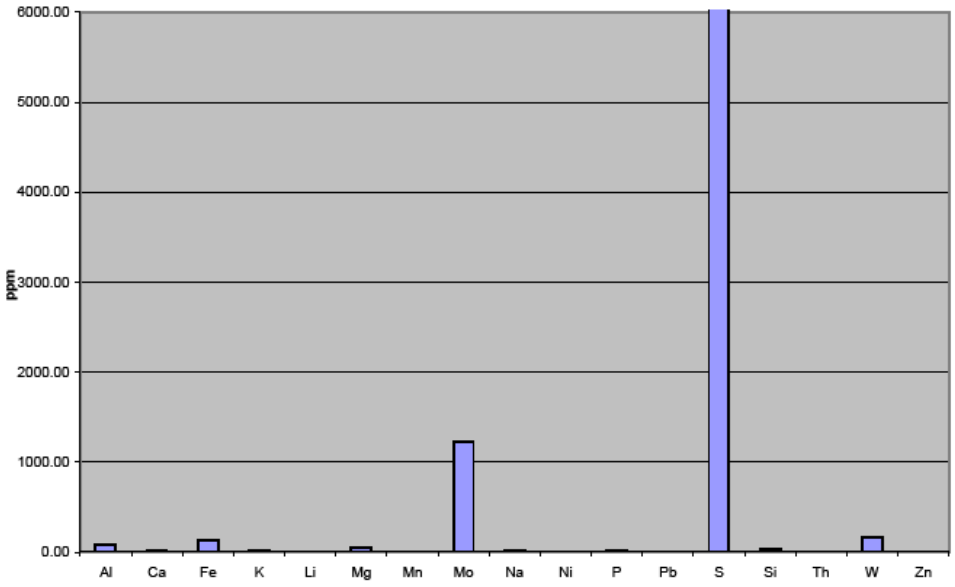


Figure 4UOCMcArthurLake

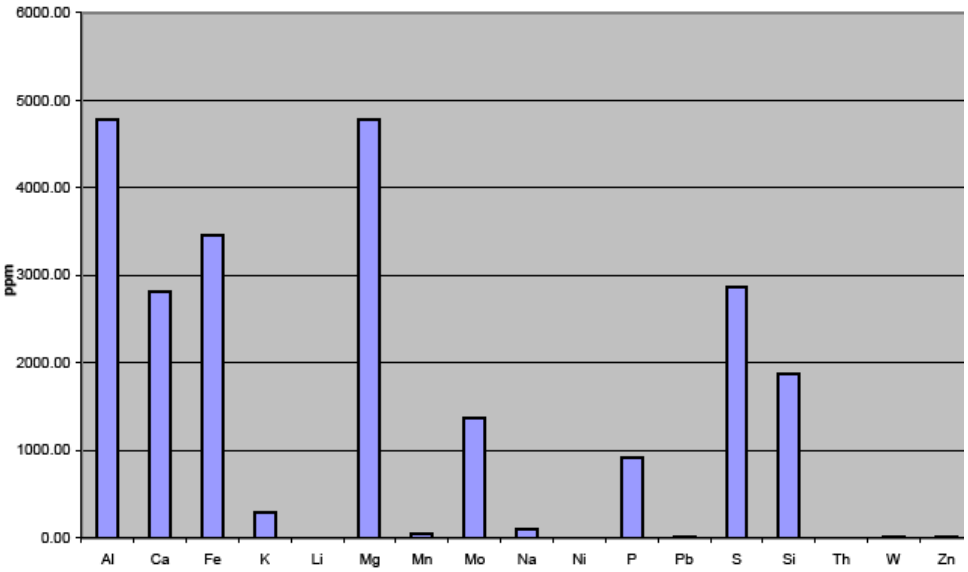


Figure 5– UOC Eldorado

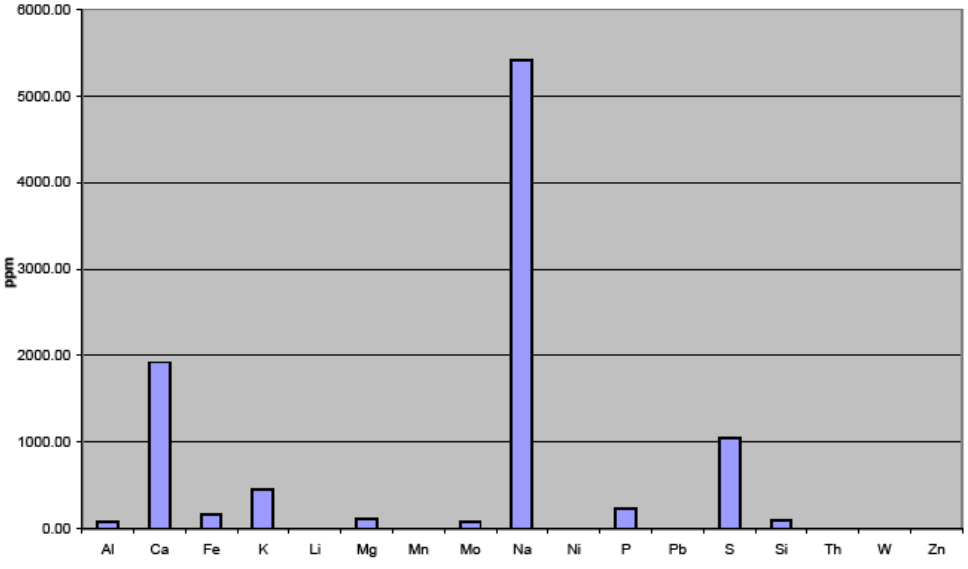


Figure 6 UOC Crow Butte

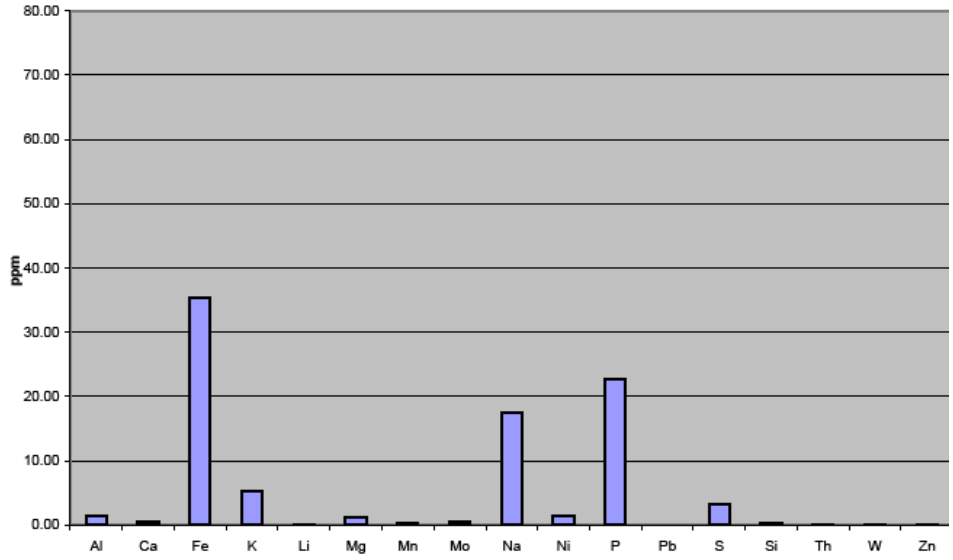


Figure 7 UO_3 product

Figure 8 shows the same impurities for an intermediary conversion product (UF_4), but using the results from a different laboratory. Surprisingly, the impurities have increased, however we can't be sure that this is not caused by differing laboratory practices. Clearly, we need to do more work in this area to ensure that we are making valid comparisons.

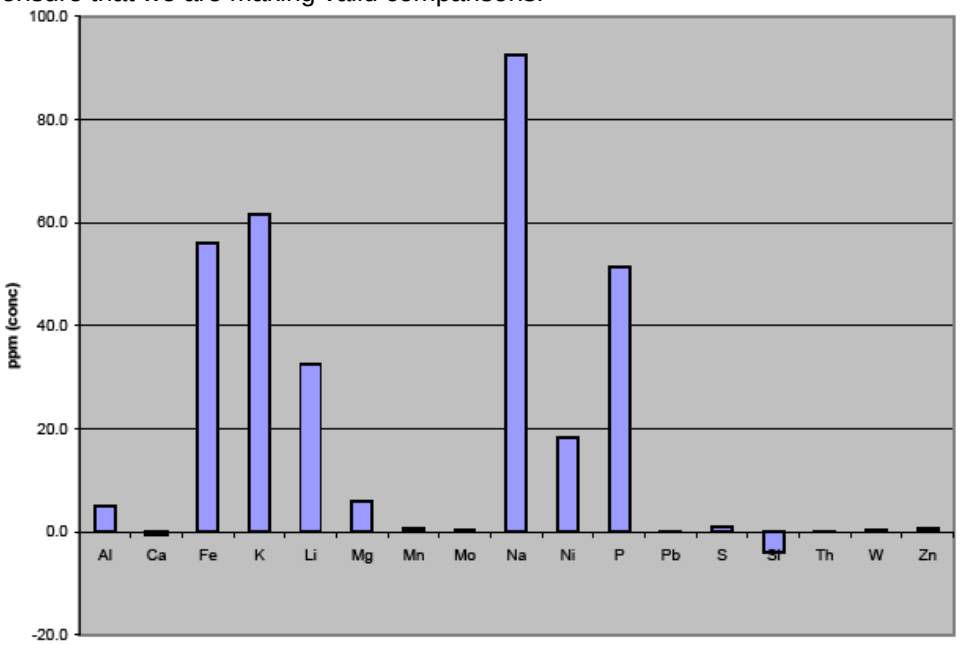


Figure 8 UF_4 (Feed1)

3.2. Isotopic data

The following table shows some of the isotopic measurements observed**.**The $^{234}U/^{238}U$ ratios clearly characterize the four UOCs. The $^{18}O/^{16}O$ ratios are also related to the UOC, but further review would be needed to confirm if this strongly related to the location of the mill where the UOC is produced. The oxygen isotope ratios for the uranium oxides also show a change.

Changes in the delta $^{235}U/^{238}U$ were also observed and are a result of redox reactions that occur during processing of the materials.

	UOC McAr	UOC Eldorado	UOC McCl	UOC Crow	UO_3	UO_2
$^{234}U/^{238}U$ per mil	28.18	2.64	13.71	-6.31	21.3	19.5
$^{235}U/^{238}U$ per mil	-1.17	-1.36	-1.03	-1.44	-1.0	-0.9
$^{18}O/^{16}O$ per mil	-3.0	-5.7	-0.4	-6.2	0.2	0.5

The oxygen and uranium isotope ratios are shown graphically in Figure 9.

Meteoric water from the Athabasca area has a lower delta ^{18}O value than that from more southern locals. Additionally, when UO_3 forms, the isotopic composition of the oxygen in the mineral is not the same as that in the water (fractionation factor). The fractionation factor for UO_3 results in UO_3 having a delta ^{18}O value about 8 per mil greater than the water. The differences between oxygen ratios for the UOCs and the UO_3 and UO_2 are a result of both the fractionation factors and the difference between meteoric water at the mills (Athabaska) and that at the processing plants.

Figure 10 shows the oxygen isotopic ratios plotted against the lead ratios. Lead in UOCs is from the ores and is very radiogenic; i.e. it has low $^{207}Pb/^{206}Pb$ ratios and high $^{206}Pb/^{204}Pb$ ratios when compared with common lead (whole earth lead) which has a $^{207}Pb/^{206}Pb$ ratio of about 0.9 and $^{206}Pb/^{204}Pb$ ratio of about 16, very similar to the lead in the grit blast. All of the process samples after UOC have lead isotope ratios closer to common lead, probably from mixing of lead from the grit blast, about 1000ppm,with that from the UOC, less than 1 ppm for most.

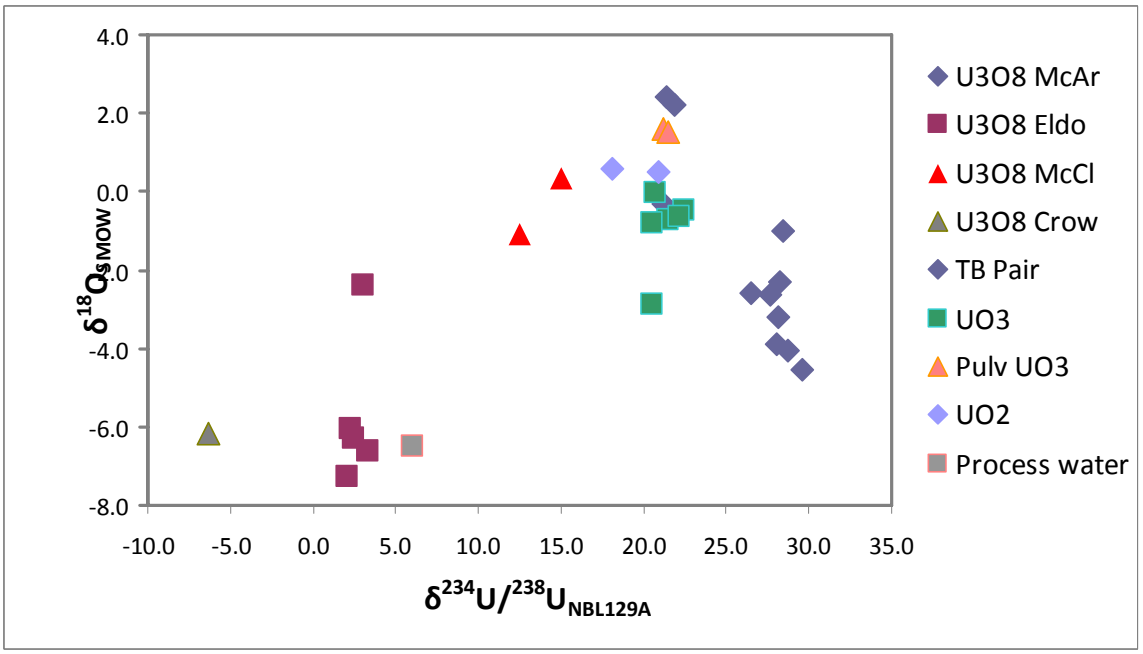
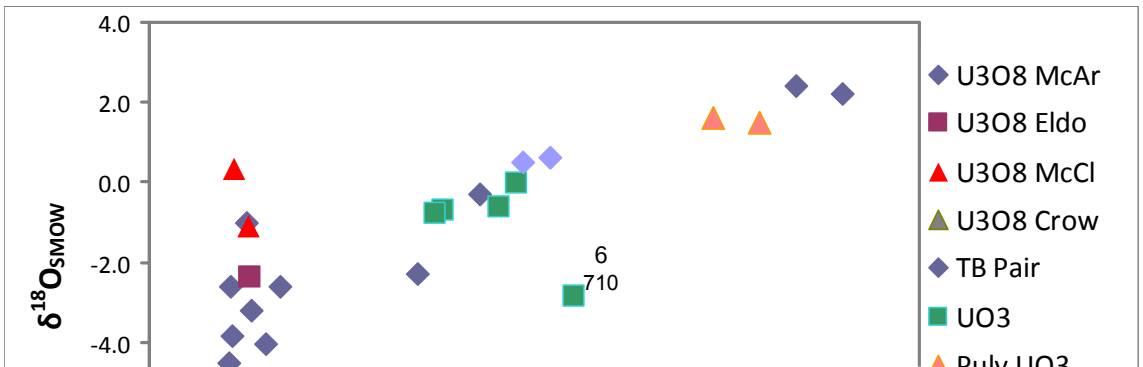


Figure 9 - Oxygen vs uranium isotopic ratio plot



4. Future directions

As indicated by earlier work, impurity measurements can be used to characterise the origins of various UOCs. Oxygen and lead isotopes will also help in this characterisation.

The variance in results for trace elements from different laboratories needs to be understood before more definitive results can be offered. This will particularly important if we are learn more about trace element changes that occur as the material passes through the refining and conversion.

Isotopic data shows promise for localising the site of a process, but the data needs further review before making firm conclusions.

As yet, no further sample collection is being planned. Future exercises may involve collection of more UOC samples to extend and confirm the data we already have. Also, should we find a basis for changes occurring due to process, we may want to repeat some part of the original sampling to confirm our theory.

5. Acknowledgements

This study has been funded through the Canadian Safeguards Support Program and the assistance of Queen's University facility for Isotope Research (QFIR). We are grateful for the assistance provided by Cameco, whose cooperation made this work possible.

Classification of uranium ore concentrate samples based on infrared spectroscopy

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Abstract:

In this work the applicability of Fourier-transform infrared spectrometry (FTIR) for nuclear forensic studies of uranium ore concentrates (UOC) are investigated. The technique was used for the identification of the type of uranium compound and various process-related impurities, which can give information on the production method of the material. The measured spectra were evaluated also by statistical means, using soft independent modelling of class analogy (SIMCA) technique to reveal less apparent similarities between the measured UOC samples.

Keywords: Nuclear forensics, Fourier-transform infrared spectroscopy, origin assessment, SIMCA, uranium ore concentrate.

1. Introduction

Illicit trafficking of radioactive, especially nuclear materials, poses a serious threat to the international community. Such materials, not only due to their radiotoxicity, but also as a result of nuclear proliferation concerns, can cause considerable risk if they are diverted and get out of regulatory control. As it is of crucial importance to obtain information on the origin and intended use of these materials for the legal procedure and to reinforce protective measures, a new scientific discipline – nuclear forensics – has evolved in the last decade [1-3].

Uranium ore concentrate (UOC), commonly known as *yellow cake*, is a generic term, which refers to the intermediate product of the uranium nuclear fuel cycle. These materials are produced (milled) by various metallurgical methods from uranium ores or recovered as a by-product of other products [4]. Typically, a uranium ore concentrate contains a high fraction of natural uranium (above 65 mass% uranium), and can involve a wide range of chemical compounds, such as ammonium uranate (AU), sodium diuranate, uranyl hydroxide, uranyl peroxide or it can be in oxide form, if the material has been calcined. They are typically obtained (recovered) by precipitating uranium after the purification by solvent extraction or ion exchange. The precipitation is realized using a variety of precipitants. The method of choice depends on price, availability of chemicals, purity required or metallurgical consideration (e.g. filtration properties of the product or the necessity to match the rest of the process).

Several parameters (indicators) can be used for the origin assessment of unknown materials in nuclear forensics, such as composition, trace elemental impurity pattern, uranium, lead or strontium isotopic composition [1,2,5,6]. These parameters can give information on the source of the feed material used for uranium production and/or the metallurgical process used for uranium milling. Several of these indicators have been extensively investigated in the last years for nuclear forensics purposes. Less attention, however, was paid to the molecular spectroscopic studies of these compounds, such as infrared spectroscopy or X-ray diffraction. These techniques can be applied not only to identify the chemical compound, but they are also capable to detect certain impurities, which are indicative to the hydrometallurgical process applied.

The aim of the present study was to investigate the possibility of Fourier-transform infrared spectrometry (FTIR) for the identification of uranium ore concentrates and to find a correlation

between the measured spectra and the metallurgical processes used. The second stage of the study was to develop classification models, which are able to group uranium ore concentrates based on their composition and impurities by the combination of mid-IR spectral information of the samples. In particular the application of supervised classification techniques was studied.

2. Experimental

2.1. Instrumentation

The FTIR measurements were performed in solid form using the KBr pellet technique. The KBr pellets of UOC samples were prepared from approximately 100 mg spectral grade KBr and 1 mg of UOC. The mixture was homogenized and ground in agate mortar, thereafter was pressed into a pellet at 600 MPa for 8 minutes. Prior to use, the KBr was heated at 100 ^{o}C and stored in a desiccator. All infrared spectra were measured in the range of 400 – 4000 cm^{-1} using a Perkin Elmer System 2000 FTIR spectrometer (Perkin Elmer Ltd., Beaconsfield, UK).

2.2. Investigated samples

Approximately 70 yellow cake samples were investigated in this study representing the world-wide UOC production. For most samples detailed information was available on the production methodology and the final product of the respective process [4,7,8]. Additionally, a reference sample of ammonium uranyl carbonate (AUC) was prepared in the laboratory by dropwise addition of 25% ammonium carbonate solution to a pure uranyl nitrate solution [9].

2.3. Data analysis

The discrimination of UOC samples was achieved using soft independent modelling of class analogy (SIMCA). Data analysis was performed using multivariate statistical analysis methods with SIMCA software (Umetrics, Sweden). For the data analysis whole FT-IR spectra of the samples were used. The range between 400–4000 cm^{-1} corresponds to 7201 wavenumbers.

3. Results and discussion

3.1. Identification of uranium ore concentrates

The most common UOCs and the observed absorption bands of the respective functional groups are shown in Table 1.

	Functional group and observed absorption frequency (cm^{-1}) [a]										
	ν(OH)	ν(NH)	δ(H_2O)			ν(UO_2)			ν(UO) and/or ν(UN)		
Ammonium uranate (AU)	3450 s, br	3170 s, br	1620 m		1400 s, br	895 s,br			470 s		
Uranyl hydroxide	3450 s, br		1620 m			890 s,br			450 s		
Sodium diuranate	3400 s, br		1620 m			830 s,br	750 m	540 m	450 s		
Uranium peroxide hydrate	3470 s, br	3140 s, br	1620 s			920 s,br	860 m		490 s		
Ammonium uranyl carbonate	3450 s,br	3140 s, br	1620 s	1520 m	1400 s,br		895 m		490 s		
α-U_3O_8	3400 w		1620 w				740 m		525	490	455

[a] w = weak, m = medium, s = strong, br = broad

Table 1: The observed IR wavenumbers of the investigated UOC materials

Ammonium uranate (AU) is one of the most common yellow cake compounds. It does not have a well-defined stoichiometry, its composition varies according to the precipitation conditions. The accepted formula is $(NH_4)_2U_2O_7$, although $UO_3.xH_2O.yNH_3$ better describes the composition, where *x* and *y* indicate the variable amount of H_2O and NH_3 in the material. AU precipitation is mostly carried out with gaseous ammonia at elevated temperature. The infrared spectrum of AU compounds have several important features (Fig. 1): two overlapping bands at 3450 and 3170 cm^{-1} arising from the O-H and N-H stretching vibrations; a band due to the vibrational bending mode of H_2O (δ_{H2O}) at 1620 cm^{-1}, its absorption is proportional to the amount of coordinated water molecules; an absorption band at 1400 cm^{-1} from the symmetric deformation of NH_3; at 895 cm^{-1} is the asymmetric stretching frequency (ν_3) of the uranyl group, its frequency increases as the NH_3 content decreases [10]; OH deformation band appears with weak intensity in the 700-1000 cm^{-1} region depending on the water content and a strong band at 470 cm^{-1}, which was assigned to the U-N stretch of an NH_3 group coordinated to uranium [11,10].

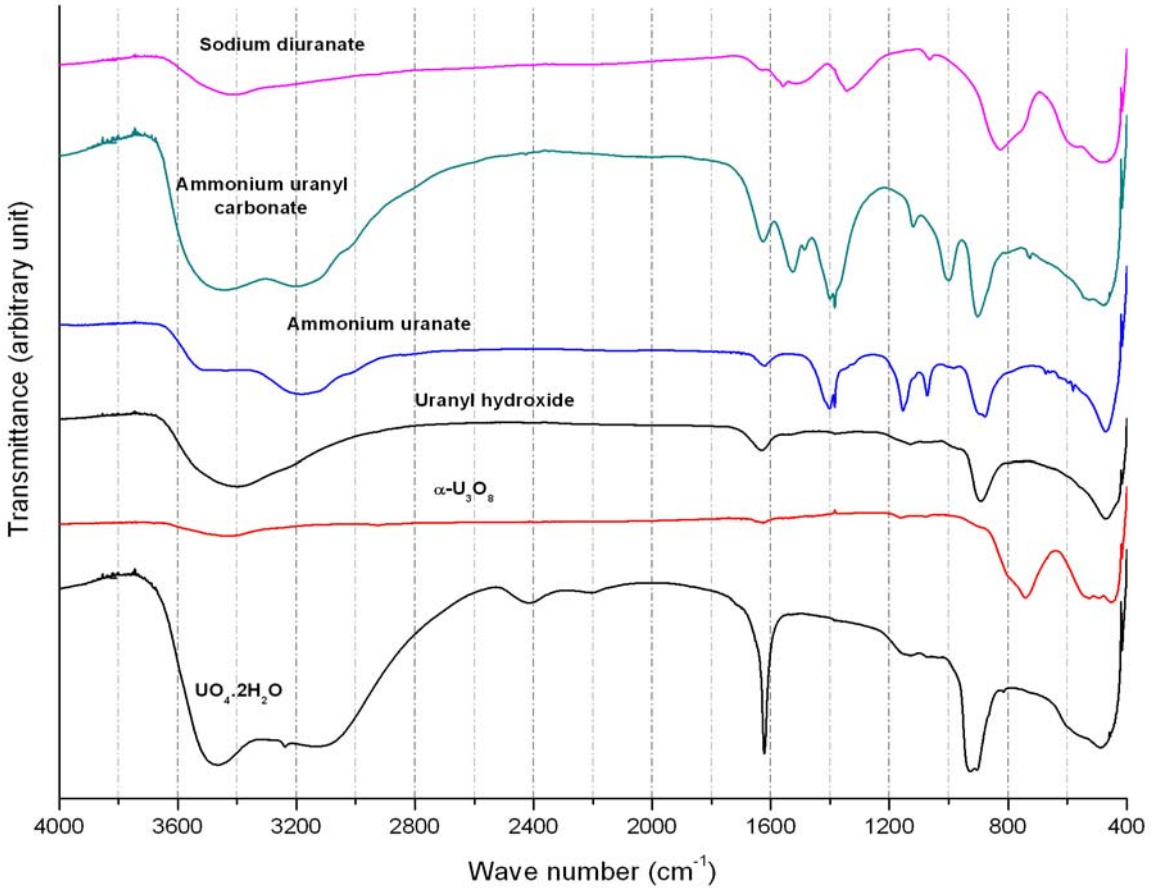

Figure 1: Typical FTIR spectra of various UOC compounds

Uranyl hydroxide or UO_3 hydrates ($UO_2(OH)_2$ or $UO_3.xH_2O$) is an orange-yellow material. It is formed when uranium is precipitated from oxidized liquors at near neutral pH. Due to its colloidal form it is difficult to filter. Most often MgO is used for precipitation at a pH of between 7.0 and 7.5. The major regions in the FTIR spectrum: a broad band at 3450 cm^{-1} due to the O-H stretching vibrations of hydroxyl-groups; a band with varying intensity due to the vibrational bending mode of H_2O (δ_{H2O}) at 1620 cm^{-1}; at 890 cm^{-1} is the asymmetric stretching frequency of the uranyl group; OH deformation band appears with weak intensity in the 700-1000 cm^{-1} region depending on the water content; a band at around 450 cm^{-1}, which was suggested to be the U-O stretching [11,10].

Sodium diuranate ($Na_2U_2O_7{\cdot}6H_2O$) is precipitated at relatively high pH (above 11) by sodium hydroxide. As the NaOH is usually used in excess, polyuranates are preferably formed. Detailed study on the vibrational spectrum was described in details by Volkovich et al. [12]. Usually it does not

contain high amount of sulphate compared to the uranyl hydroxide. The major regions in the FTIR spectrum: a broad band at 3400 cm^{-1} due to the O-H stretching vibrations of hydroxyl-groups and coordinated water molecules; a band with varying intensity due to the vibrational bending mode of H_2O (δ_{H2O}) at 1620 cm^{-1}; a strong and broad band at 830 cm^{-1} is the asymmetric stretching frequency of the uranyl group with a smaller peak (shoulder) around 750 cm^{-1}; a medium intensity peak at 540 cm^{-1}, which appears as shoulder can be assigned to the U-O stretching; OH deformation band appears with weak intensity in the 700-1000 cm^{-1} region depending on the water content; a band at around 450 cm^{-1}, which was suggested to be the U-O stretching [12,13]. As this compound is often the final product if alkaline carbonate is used for the ore leaching and stripping after purification, the carbonate bands often appear at 1520 cm^{-1} overlapping the water peak. Further verification of the compound can be done by the measurement of Na and U-content.

Hydrogen peroxide as a precipitant became widely popular in the last decade. It does not only eliminate the environmental problems related to ammonia, but also provides a more pure product. Uranium is precipitated as uranium peroxide hydrates with H_2O_2 at a relatively low pH of 3.5. The product exists in two crystalline forms, $UO_4.4H_2O$ and $UO_4.2H_2O$ depending on the precipitation temperature [14]. The major regions in the FTIR spectrum: two overlapping broad bands at 3470 and 3140 cm^{-1} due to the O-H stretching vibrations; often a sharp peak at 3330 cm^{-1} from the OOH stretching is also visible; a band with high intensity due to the vibrational bending mode of H_2O (δ_{H2O}) at 1620 cm^{-1}; a strong band at 920 cm^{-1} is the asymmetric stretching frequency of the uranyl group, sometimes with a shoulder at 860 cm^{-1} from the stretching of the peroxo-group; a band at around 490 cm^{-1} from the U-O stretching [14-16]. The FTIR spectra of the two hydrates are different, which can be used to distinguish them by the OH absorption bands and the shape of the δ_{H2O} peak: in case of the dihydrate the band at 3450 cm^{-1} is stronger than the one at 3200 cm^{-1}, while the H_2O deformation appears as a sharp peak at 1620 cm^{-1} [14]. In most cases $UO_4.2H_2O$ was found among the investigated uranium peroxide samples.

Ammonium uranyl carbonate (AUC) is a less frequent form of uranium ore concentrates, however, it is an important intermediate product for UO_2 production. Usually AUC is precipitated from uranyl nitrate or dilute carbonate solution with ammonium carbonate. The major regions in the FTIR spectrum: two overlapping bands at 3450 and 3170 cm^{-1} arising from the O-H and N-H stretching vibrations; a band at 1620 cm^{-1} due to the vibrational bending mode of H_2O (δ_{H2O})**;** a broad absorption band at 1400 cm^{-1} from the symmetric deformation of NH_3; the antisymmetric stretching of CO_3^{2-} at 1520, 1050, 720 and 690 cm^{-1}; strong band at 895 cm^{-1} is the asymmetric stretching frequency of the uranyl group; a band at around 490 cm^{-1} from the U-O stretching [17,18].

Various uranium oxides can also belong to uranium ore concentrates. Calcination is used to increase the grade of the concentrate and to remove certain impurities, such as ammonia or sulphate. The temperature used depends on the impurities to be removed, however, it is usually less than 700 ^oC to avoid the formation of components of low solubility. The final product primarily depends on the initial material compound and the temperature used for calcination. For instance, the calcination of ammonium uranate results in β-UO_3 with varying amounts of water and ammonia if the temperature is below approximately 400 ^oC, and is converted to α-U_3O_8 at about 600-700 ^oC. Using uranium peroxide as starting material the intermediate product is α-UO_3 if the calcination is done at lower temperatures (below 250 ^oC), which converts to α-U_3O_8 at about 650 ^oC [19]. Thus the exact compound type can give information on the possible intermediate product before calcination as well as a rough estimate on the calcination temperature. A summary on the FTIR measurements of various uranium oxides was published by Allen and Holmes [20]. In case of the oxide UOC samples mostly α-U_3O_8–form was found. The FTIR spectrum of this compound is relatively simple: a small peak at 3400 cm^{-1} is due to the O-H stretching vibrations of hydroxyl-groups as a result of the remaining moisture content of the sample; a band with small intensity at 1620 cm^{-1} is from the vibrational bending mode of H_2O (δ_{H2O}); a strong band at 740 cm^{-1} is the asymmetric stretching frequency of the uranyl group; a strong, complex absorption band, which contains three peaks at 525, 490 and 455 cm^{-1}.

Several impurities can also be identified in the FTIR spectra. These detected impurities are indicative to the pregnant liquor composition, from which uranium is precipitated. Their amount depends highly on the precipitation conditions and the effectiveness of the occurrent washing step. Although FTIR is

less sensitive than other methods, such as ion chromatography [21], it offers a rapid identification of several (anionic) impurities. The most typical impurities identified are sulphate, nitrate and carbonate. Sulphate, which is widely used for the leaching of the ore as well as in the forthcoming purification steps (e.g. for uranium elution in ion exchange separation), is often present in the FTIR spectrum. It has a well-detectable peak at 1120 cm^{-1}. Nitrate gives a very sharp characteristic absorption band at 1384 cm^{-1}. Although nitrate is highly soluble and can be easily eliminated by subsequent washing steps, it was still measureable in several yellow cakes, where nitrate solution is used for uranium stripping after the ion exchange purification. Carbonate is often present in sodium diuranate samples, as sodium hydroxide is frequently applied for precipitation from alkali solutions. FTIR offers a rapid measurement tool for carbonate detection in contrast to ion chromatography, as it is uses most often dilute carbonate solution as eluant. The most intense carbonate peaks appear at 1330 cm^{-1} and 1550 cm^{-1} (carbonate antisymmetric stretching region), and a medium intense peak at 1050 cm^{-1} (carbonate stretching) [18].

3.2. Classification analysis technique: Soft Independent Modelling of Class Analogy (SIMCA)

Soft independent modelling of class analogies (SIMCA) is a well-known supervised classification technique [22,23]. In SIMCA, each class model existing in a training set is defined by principal component analysis (PCA) [24], which is generally based on cross validation. Once the principal components of each class model are calculated, the critical limits of each class in the space are defined by a critical value of the distance towards the model. Based on the critical limits, new samples can be evaluated and considered either as outlier and rejected by the class models or accepted and recognized as being part of the class. SIMCA is more focused on the similarities between the samples of the same class than dissimilarities between the classes.

For the interpretation of the results, Cooman's plot is a useful tool. It is generally applied for the discrimination of two classes and obtained by plotting the distances of class 1 model against to the distances of class 2 model (Fig. 2). As it can be seen from the Fig. 2, there are four different zones defined by the critical limits and each zone in the space defines the location of the samples. There are two axes representing the distance of an individual sample from the model. Two classes models are plotted against each other with the critical levels as straight lines which shows the boundaries to the classes region. Generally these two areas are the corresponding area for the two classes. Any sample whose distance is larger than the critical distance can be classified as not belonging to the class.

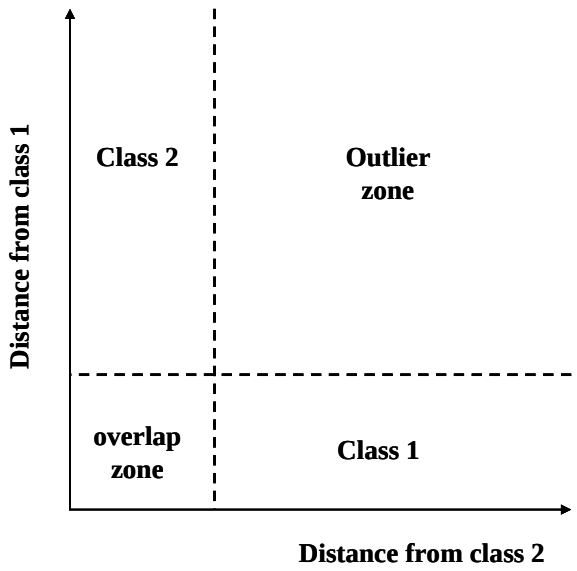

Figure 2: The Cooman's plot

3.3. Classification of the measured uranium ore concentrate (UOC) samples

A classification model for uranium ore concentrates (UOC) based on their composition was developed. Three major classes could be identified using the model at the first stage. The denomination of classes was based on the major composition of the materials. The classes were named: sodium diuranate (SDU), identified as Class 1; ammonium uranate (AU), identified as Class 2; and a complex group including peroxides, oxides and hydroxides identified as Class 3 (OX). All the uranium ore concentrate samples were divided randomly into two parts: training and test set. The training set including three different classes contains 9, 17 and 32 samples for each class, respectively. An independent test set including 9 samples was used to check the prediction capability of the class models. Prior to PCA modelling to each class, standard normal variate (SNV) transformation was applied to the training set. SNV is a mathematical transformation method that is used to remove the slope variations, baseline shifts and to correct scattering effects. Each spectrum is corrected individually by first centering the spectral values and then the centred spectrum is scaled by the standard deviation value which is calculated from the individual spectral values. Mean centering has been done on the result spectra by calculating the average of the spectrum of the data set and subtracting the average from each spectrum.

PCA modelling was performed for each class present in the training set, separately. The model is generally based on leave-one-out cross validation at the 95% confidence level. Two PCA models which belonged to the two different classes were used to construct to Cooman's plot (Fig. 2). Principal components (PCs) of 3 class models were calculated independently and overall statistics of PC class models for each multivariate data set is shown in Table 2.

PCA class models	Number of PCs	R^2 *X* (cum) (%)
Class 1 (SDU)	4	72.4
Class 2 (AU)	8	90.3
Class 3 (OX)	15	97.5

Table 2: PCA class models and statistics for UOC of mid-IR spectral information

Class 2 and Class 3 were preferably to be plotted against each other in Cooman's plot, since the corresponding value of explained variance for these two classes has the highest value. As Fig. 3 reveals, each sample of Class 2 and Class 3 was correctly plotted in its critical limits apart from the origin, which underlines the discrimination of these two classes based on UOC samples in the training set. Class 1 samples were plotted in the outlier zone in this representation, i.e. distinctly separated from the Class 2 and Class 3 samples. It is also noteworthy that Class 2 (AU) samples form a well-separated group, while Class 1 (SDU) and Class 3 samples (peroxides, oxides and hydroxides) are more alike. This is due to the chemical similarity (and by this means also the similarity in the mid-IR spectra) between the SDU, hydroxides, oxides and peroxides. Based on the classification result, the most contributed mid-IR region to the each class models was investigated and a significant contribution for the ammonium uranate samples was found between 1200 – 1600 cm^{-1} wavenumbers, corresponding to the high contribution of the ammonium peak. For the sodium diuranate samples the most contributing mid-IR region is between 1100–1500 cm^{-1} wavenumbers (mainly from carbonate and sulphate impurities), whereas the regions between 400–1600 cm^{-1} and 3200–3600 cm^{-1} wavenumbers are the significant regions for the classification of UOCs samples including hydroxides, oxides and peroxides (corresponding to the stretching of uranyl group, U-O and O-H stretching). For Class 2, one sample (shown in Fig. 3 in Class 1 directly at the border of Class 1 and Class 2 zone) out of 17 samples (94.12% correct assignation) is placed outside the limit of each membership, while for Class 3 two samples (marked with asterisk in Fig. 3 at the boundary of Class 1 and Class 3 in the Class 1 zone) out of 32 samples placed in outer of the limit of each membership, which is the region of Class 1 membership (93.75% correct assignation).

In case of the misassigned samples, the SIMCA analysis revealed that it is the elevated level of sulphate impurity at 1040–1110 cm^{-1}, which caused the slight shift of the samples towards the outlier zone. These results show that uranium ore concentrates that contain more than one impurity, will be correctly assigned (the composition can be identified), however, they may tend to be grouped to the outlier zone due to the presence of the additional feature in the mid-IR spectrum. Using SIMCA, these regions of interest can be readily found making the identification of the impurity easier.

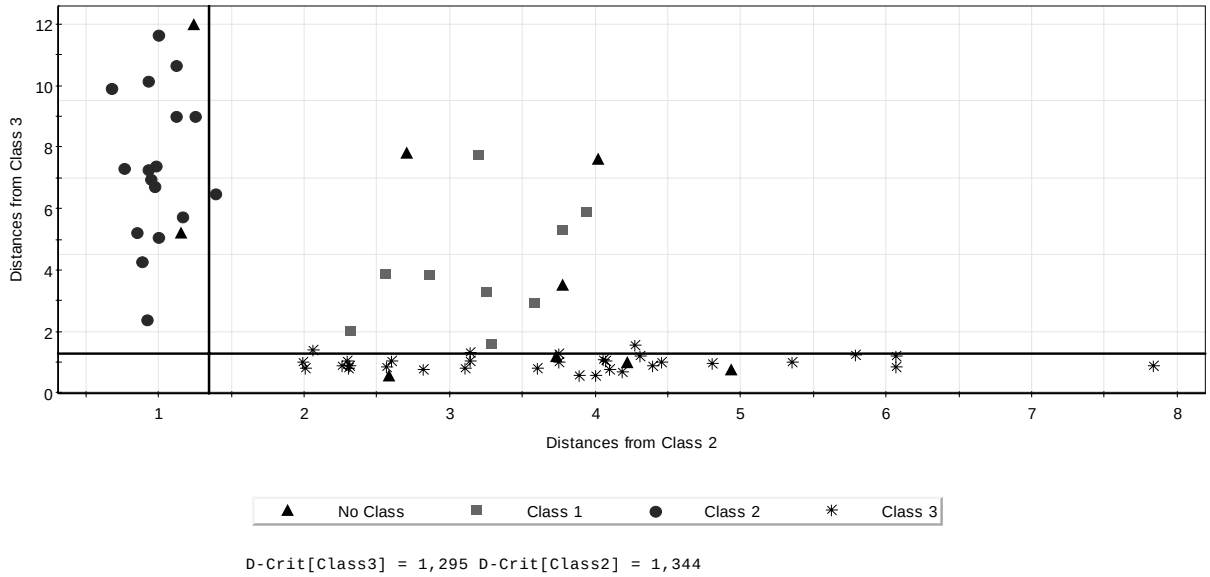

Figure 3: Cooman's plot for the classification of uranium ore concentrates using mid-IR spectra: ▲ test set (denoted as no class in the software) ■ Class 1 (sodium diuranate) • Class 2 (AU) ∗ Class 3 (peroxide, oxide and hydroxide)

The class models were also tested using an independent test set. These samples were chosen randomly among the uranium ore concentrate samples and were predicted by the help of the class models. According to the results the prediction ability of the class models is 77.8%. Two test samples from Class 2 and Class 3 (out of 9 samples) were assigned incorrectly to the Class 1 membership. The contribution plot of the samples proved that the misassigned samples contain either elevated level of sulphate or are partially calcined AU sample. In such cases the samples will be grouped to the outlier zone indicating the presence of an additional component to the spectrum.

4. Conclusions

FTIR spectrometry was found to be an easy and simple tool to identify the composition of uranium ore concentrates. The method is almost non-destructive and uses a small amount (a few mg) for the analysis. Besides the actual chemical composition, several anionic impurities can be identified in the mid-IR spectrum, which can give clues on the production method. In this study the efficiency of FTIR in combination of SIMCA for the differentiation of uranium ore concentrate class models was demonstrated. Results suggest that application of SIMCA to mid-IR spectral information of UOC is in most of the cases successful for the classification of UOC samples.

5. Acknowledgement

The authors wish to acknowledge the UK Support Programme to IAEA for providing samples of yellow cake. Similarly, we would like to thank also the IAEA providing additional samples. The support provided by the Australian Safeguards and Non-proliferation Office (ASNO) for the acquisition of samples from Australia is highly appreciated.

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Uncertainty assessment in gamma spectrometric measurements of plutonium isotope ratios and age

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Abstract

A method for the assessment of the combined uncertainty in gamma spectrometric measurements of plutonium composition and age was evaluated. Two materials were measured. High resolution inductively coupled plasma mass spectrometry (ICP-SFMS) was used as a reference method for comparing the results obtained with the gamma spectrometric method for one of the materials. For this material the measurement results were in agreement between the two methods for all measurands. Moreover, the uncertainty in all isotope ratios considered ($R_{Pu238/Pu239}$, $R_{Pu240/Pu239}$, $R_{Pu241/Pu239}$, and $R_{Am241/Pu241}$ for age determination) were limited by counting statistics.

Keywords: plutonium, isotope ratio, composition, age, gamma spectrometry, uncertainty

Nuclear Forensics cooperation in the European Union: Results of the joint analysis of natural uranium samples of Hungarian origin

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Abstract:

Illicit trafficking of nuclear and other radioactive material is a subject of serious concern due to the radiological hazard and the proliferation risks associated with such material. Nuclear forensics is a scientific discipline interfacing law enforcement, nuclear science and non-proliferation. Through nuclear forensic analysis, information on the history and on the potential origin of intercepted nuclear material can be obtained by investigating the characteristic parameters of such material.

Substantial support to the national authorities of EU Member States has been provided in more than 35 cases through nuclear forensic analysis performed at the Institute for Transuranium Elements (ITU). In addition to the technical challenges, also the organizational provisions have to be in place to facilitate an effective and efficient nuclear forensics support. A mechanism for providing nuclear forensics support has been described in IAEA Nuclear Security Series No. 2. Appropriate arrangements are in place between the European Commission, more specifically at the Institute for Transuranium Elements, and some EU member states. The paper illuminates the nuclear forensics methodology that is used in the analysis of seized nuclear material. This methodology has been exercised in so-called "joint analysis". In a recent undertaking, samples of nuclear material were transported from Institute of Isotopes (IoI, Hungary) to ITU and were analyzed in both institutions.

Discussion of the results obtained during the joint analysis for characteristic parameters (such as isotopic and elemental composition, macroscopic and microscopic appearance, age, IR spectrum) is described in the paper.

Keywords: nuclear forensics, uranium, ICP-SFMS

1. Introduction

Illicit trafficking of nuclear and other radioactive materials has emerged as a truly international problem and is indeed dangerous threat to international security. The problem of theft or loss of nuclear materials has affected countries on all five continents proving once again the international scale of this issue. Concerns about nuclear smuggling resulted in the development of a new field known as nuclear forensics [1].

The "birth" of the nuclear forensics can be considered being two decades ago after the break-up of the former Soviet Union, when smuggling incident of nuclear material (uranium oxide pellets, metallic uranium, uranium ore concentrates, etc.) started to occur frequently, especially in Central Europe. In the beginning the methods and techniques used in nuclear forensics analysis were adapted from other

analytical fields, such as nuclear safeguards and geology, but during the last decade the nuclear forensics has evolved and matured to an own sophisticated discipline.

Nuclear forensics is the analysis of nuclear materials recovered from e.g. seizure of fresh nuclear materials to provide evidence for determining the history of the sample material. It contributes significantly to the identification of the sources of the materials and the industrial processes used to obtain them [2]. Therefore, nuclear forensics is not only an essential tool to provide evidence for the prosecution of the illicit trafficking case, but also for the prevention of further trafficking from the same source.

Measurement of certain characteristic parameters of nuclear materials of unknown origin can be utilized for their identification and characterization. Such useful analytical parameters for identification of nuclear material include isotopic composition of uranium (and plutonium), morphology features (e.g. surface roughness), age or production date and impurity spectrum (e.g. rare earth elements and other elements in specific pattern as a fingerprint) [3-7]. As an example, impurities, especially the rare-earth elements, are present in nuclear materials in widely variable concentrations due to different reasons, e.g. some lanthanides may be added to the nuclear materials as burnable poisons (Er, Gd) in order to control the reactivity of the fuel in nuclear reactors. Rare-earth elements can also be present in trace-level amounts either as residues from the raw material or as a contamination of the process [4]. Therefore, analysis of this parameter is particularly informative. Another very important characteristic is the age of the material.

Nuclear forensics activities are performed at the ITU since the beginning of 1990's. These activities include analysis of seized samples (up to now 39 full nuclear forensics investigations have been performed), R&D, training and close cooperation with institutions of different EU member states and worldwide. Such cooperation has been established also between ITU and the Institute of Isotopes (Hungary) and this was exercised now for the second time in the frame of "joint analysis". The first exercise took place in 2006 and it included analysis of three different uranium pellets seized in 1990's in Hungary. The material analyzed in the present exercise was a uranium ore concentrate sample from Hungarian origin.

The aim of this paper is to discuss the results obtained from analyzed characteristic parameters (such as isotopic and elemental composition, microscopic appearance) of the Hungarian samples.

2. Experimental

2.1. Instrumentation

Impurity measurements were carried out at both institutes using a double focusing magnetic sector inductively coupled plasma mass spectrometer (ICP-SFMS) equipped with a single electron multiplier (ELEMENT2, Thermo Electron Corp., Germany). The ICP-MS instrument is attached to a glove box at ITU.
Prior to the sample analysis the instrument was tuned using a 1 ng g^{-1} multielemental solution (Merck, Darmstadt, Germany). The optimization was carried out with respect to maximum uranium sensitivity and low UO^+/U^+ ratio. The sensitivity was approximately 1.4×10^6 cps for 1 ng g^{-1} ^{238}U and the UO^+/U^+ ratio ~ 3.5×10^{-2}.

The gamma spectrometric analysis of uranium ore concentrate was performed using a planar and a coaxial HPGe detector at ITU. The coaxial detector was installed in a low background environment. The detection limit for ^{232}U was obtained from the spectrum taken with the coaxial detector, using the peaks of ^{232}U daughters, as well as a relative efficiency curve constructed from the peaks of ^{238}U daughters.

An aliquot of the sample stock solution was diluted in 2M HNO_3 to a concentration of about 80 μg U/ml and the isotopic composition of uranium was measured by TIMS (Finnigan, MAT261) using the total evaporation method.

The FTIR measurements were performed in solid form using the KBr pellet technique. The KBr pellets of UOC samples were prepared from approximately 100 mg spectral grade KBr and 1 mg of UOC. The

mixture was homogenized and ground in agate mortar, thereafter was pressed into a pellet at 600 MPa for 8 minutes. Prior to use, the KBr was heated at 100 °C and stored in a dessicator. All infrared spectra were measured in the range of 400 – 4000 cm^{-1} using a Perkin Elmer System 2000 FTIR spectrometer (Perkin Elmer Ltd., Beaconsfield, UK). The transmittance measurement was done with 2 cm^{-1} resolution.

2.2. Samples

17 uranium ore and one uranium concentrate (yellow cake) samples were transported to ITU. Most of the samples originated from the main uranium ore deposit (former mining area) in Hungary that is located in the south of the country at the foot of the Mecsek Mountains. The geological cross section in the western Mecsek from 2500 m depth to the surface consists of the following minerals: sandstone, rhyolite, clay stone, Kővágószőlős sandstone formation with uranium ore-bearing layers and covered by sandstone and limestone. A wide variety of uranium minerals exist in the Mecsek Mountains: uraninite, pitchblende, soddyite, autunite, liebigite, zippeite, uranopilite and clarkeite [8]. Deposit type in Kovagoszollos: sandstone – tabular.
The uranium ore samples presented in the paper signed with codes: G7359, U-I-218, U-I-754 refer to their origin (different borings).

Other uranium ore samples originated from different uranium deposits in the country at the western border and north-eastern part of Hungary (close to Sopron, Bükk Mountains and the Lake Balaton). The samples were provided by the MecsekÖko Zrt. for the request of the Hungarian Atomic Energy Authority.

2.3. Sample preparation

Analysis of the uranium containing samples was carried out from solutions prepared by acidic digestion and dilution (at ITU) as well as from extraction chromatographic separation after the digestion (at IoI).

For the acidic digestion approximately 300–500 mg of sample was weighed into poly-propylene tube (IoI) and Teflon vial (ITU) and dissolved in 9 ml acid mixture (concentrated distilled nitric acid, ultrapure HCl and HF) while heating to 95 °C on a hot-plate for 20 h. Approximately 300 µL of this stock solution was weighed into a polyethylene vial and diluted 100 000 times using ultra-pure water in order to adjust the proper HNO_3 concentration and also decrease the HF concentration load to the ICP-SFMS. After the addition of Rh internal standard, the samples were analysed by ICP-SFMS using external calibration.

Other 300 µL portion of the stock solution was weighed into a polyethylene vial and diluted fourfold using ultra-pure water in order to adjust the proper HNO_3 concentration. This aliquot was used for the lanthanide separation. The lanthanide content of the sample aliquots was separated using extraction chromatography by the selective retention of trivalent lanthanides and actinides on the TRU™ resin in 3M nitric acid medium. After conditioning of the resin with 10mL 2M HNO_3 the sample aliquot was loaded on the column. After washing the column and removal of the non-retaining matrix components with 2mL of 2M HNO_3, the lanthanides were stripped from the column into a Teflon beaker using 1mL concentrated HCl followed by 4mL of 4M HCl. After the addition of 200 µL ultra-pure HNO_3 to the final fractions, the samples were evaporated to almost complete dryness on a hot-plate in order to destroy the organic resin residuals. The residue was dissolved in 1mL of 1% (m/m) ultra-pure nitric acid. After the gravimetric weighing of the final fraction and the gravimetric addition of Rh internal standard, the samples were analysed by ICP-SFMS using external calibration [7].

For impurity measurements the same digested and diluted solutions were used. Another method was the extraction chromatographic separation of the trace elements using UTEVA™ resin washed by 3X20 mL ultra-pure water and conditioned by 20 ml 8M HNO_3. Sample was loaded on the column and the impurities were eluated using 20 ml 8M HNO_3. The fraction was diluted by ultra-pure water in order to adjust the proper HNO_3 concentration. After the addition of Rh internal standard, the samples were analysed by ICP-SFMS using external calibration.

3. Results and discussion

3.1. Analytical results of the uranium ore concentrate (yellow cake) sample

Uranium ore concentrate sample was investigated with respect to its isotopic, elemental and molecular composition and morphology, respectively. These experiments were performed using ICP-SFMS at both institutions and some other techniques (Isotope Dilution Mass Spectrometry (IDMS), Thermal Ionization Mass Spectrometry (TIMS), High Resolution Gamma Spectrometry (HRGS) and Scanning Electron Microscopy (SEM), Fourier Transform Infrared Spectroscopy (FTIR)) available at ITU.
The isotopic composition was determined by ICP-SFMS at IoI and by TIMS and HRGS at ITU. The morphology of the sample was studied using SEM. Impurities were analyzed by ICP-SFMS.
The uranium results obtained by different techniques are summarized in Table 1.

	HRGS results (ITU)	**TIMS results (ITU)**	**ICP-SFMS results (IoI)**
		Value, weight % (k=2)	
^{232}U	$<10^{-11}$	n.d.*	n.d.*
^{234}U	0.0051± 0.0028	0.00538 ± 0.00033	0.0058 ± 0.0003
^{235}U	0.718 ± 0.022	0.71046 ± 0.00040	0.7166 ± 0.0082
^{236}U	<0.001	Bdl.**	Bdl.**
^{238}U	99.28 ± 0.02	99.2838 ± 0.0015	99.2772 ± 0.0433
U total (Wt%)	n.g.***	65.7 ± 0.1	63.5 ± 2.7

* n.d.: Not detectable
**Bdl.: Below detection limit
***n.g.: Not given

Table 1: Uranium isotopic composition results of the Hungarian uranium ore concentrate sample

The uranium isotopic composition and total uranium content of the yellow cake sample measured by different techniques at ITU (TIMS, HRGS) showed good agreement with the IoI ICP-SFMS results within the combined uncertainties. The isotopic composition of the yellow cake sample corresponds to the natural uranium composition as expected.

	Concentration in μg g^{-1} (k=2)	
	IoI	**ITU**
Ce	0.215±0.061	0.35±0.04
Dy	0.056±0.009	0.055±0.007
Eu	0.024±0.001	0.024±0.003
Er	0.041±0.004	0.044±0.005
Gd	0.1145±0.0007	0.33±0.04
Ho	0.0127±0.0006	0.014±0.002
Lu	0.0071±0.0006	0.0074±0.0009
Nd	0.042±0.006	0.042±0.005
Pr	0.026±0.003	0.010±0.001
Sm	0.027±0.001	0.016±0.002
Tb	0.0084±0.0006	0.0078±0.0009
Tm	0.0067±0.0005	0.0069±0.0008
Y	0.74±0.14	0.74±0.09
Yb	0.036±0.003	0.059±0.007

Table 2: Rare earth elements concentrations in the Hungarian uranium ore concentrate sample by ICP- SFMS

	Concentration in μg g^{-1} (k=2)	
	IoI	**ITU**
Al	50.5 ± 12.1	45±5
Ba	92.4 ± 4.2	56±7
Ca	9041 ± 344	8593±1031
Cd	0.008 ± 0.004	0.52±0.06
Co	1.67 ± 0.22	1.82±0.22
Cr	16.5 ± 1.9	16.8±2.0
Cu	37.9 ± 0.8	42±5
Fe	695 ± 19	779±93
Ga	< 0.005	< 0.03
Mg	892 ± 6	910±109
Mn	452 ± 16	466±56
Ni	6.85 ± 0.65	7.57±0.91
Pb	10.60 ± 0.02	16.9±2.0
Sr	52.1 ± 1.2	53±6
Tl	<0.07	< 0.05
Zn	40.0 ± 1.9	78±9

Table 3: Trace elements concentrations in the Hungarian uranium ore concentrate sample by ICP-SFMS

Table 2 and Table 3 show rare earth element and other element (impurities) results obtained using ICP-SFMS instrument at ITU and IoI. Data agreement within combined uncertainties is suitable in most of the cases. Differences in the concentration could be originated from inhomogeneity of the samples and different type of sample preparation was used (in one case acidic digestion and dilution only and in the other case extraction chromatographic separation).

3 examples from the 17 uranium ore samples were selected. Agreement or disagreement of rare earth element content in the samples analyzed by ICP-SFMS at ITU and IoI are presented in Table 4 and Table 5.

	Uranium Ore samples (and their codes) Concentration (μg g^{-1} REE, % U) (k=2)					
	G7359		U-I-218		U-I-754	
	IoI	ITU	IoI	ITU	IoI	ITU
Ce	3.56±0.31	3.68±0.09	268±50	221±4	16.5±3.1	19.9±0.6
Dy	0.166±0.062	0.104±0.035	23.7±3.1	20.2±0.4	0.300±0.052	0.347±0.146
Eu	0.164±0.024	0.132±0.016	7.15±1.50	9.74±0.11	0.397±0.019	0.391±0.023
Er	0.055±0.001	0.054±0.025	10.2±0.6	10.7±0.1	0.124±0.037	0.160±0.064
Gd	0.213±0.053	0.228±0.013	27.4±6.2	29.9±0.6	1.07±0.11	1.11±0.35
Lu	< 0.07	< 0.05	0.523±0.031	0.939±0.267	0.0072±0.0009	0.014±0.003
Pr	0.213±0.051	0.261±0.043	27.5±1.8	37.2±0.6	1.46±0.02	1.04±0.05
Sm	0.191±0.048	0.257±0.102	20.6±3.5	28±1	0.874±0.093	0.908±0.119
Tb	0.039±0.006	0.038±0.011	4.54±0.98	3.95±0.19	0.107±0.034	0.101±0.018
Tm	0.009±0.002	0.011±0.003	1.16±0.05	1.29±0.14	0.0069±0.0008	0.021±0.009
Y	1.08±0.11	1.02±0.13	60±8	117±1	2.47±0.25	4.21±0.09
Yb	0.05±0.01	0.065±0.031	5.92±1.12	7.65±0.42	0.05±0.02	0.091±0.041
U (Wt%)	3.61±0.41	3.16±0.14	30.6±5.1	35.4±1.7	3.31±0.32	3.11±0.06

Table 4: Concentrations of rare-earth elements and U in the Hungarian uranium ore samples analyzed by ICP-SFMS (Ore samples originated from Mecsek region of Hungary, former uranium ore mine)

The measured REE pattern in the uranium ore concentrate and in the uranium ore samples is shown in Figure 1 and Figure 2. The REE concentrations are normalized to the chondrite values of Anders and Grevesse [9] for better comparison.

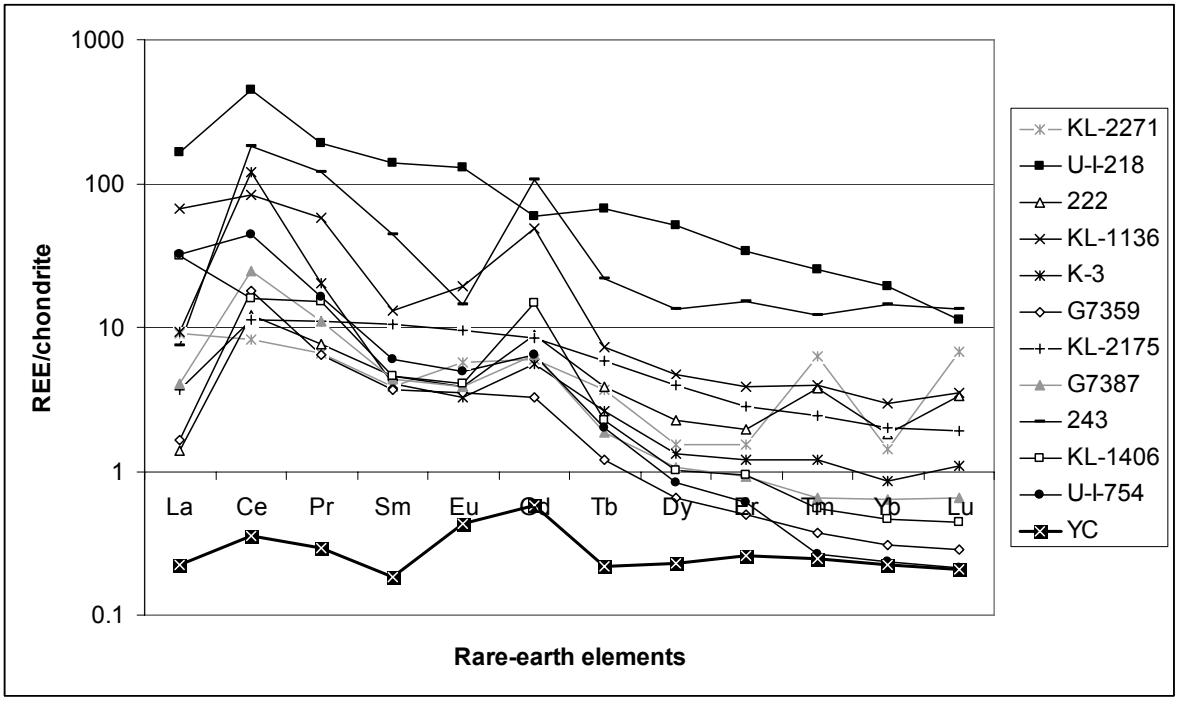

Figure 1: REE pattern of the yellow cake and the uranium ore samples originated from the Mecsek Mountains (normalized values to chondrite)

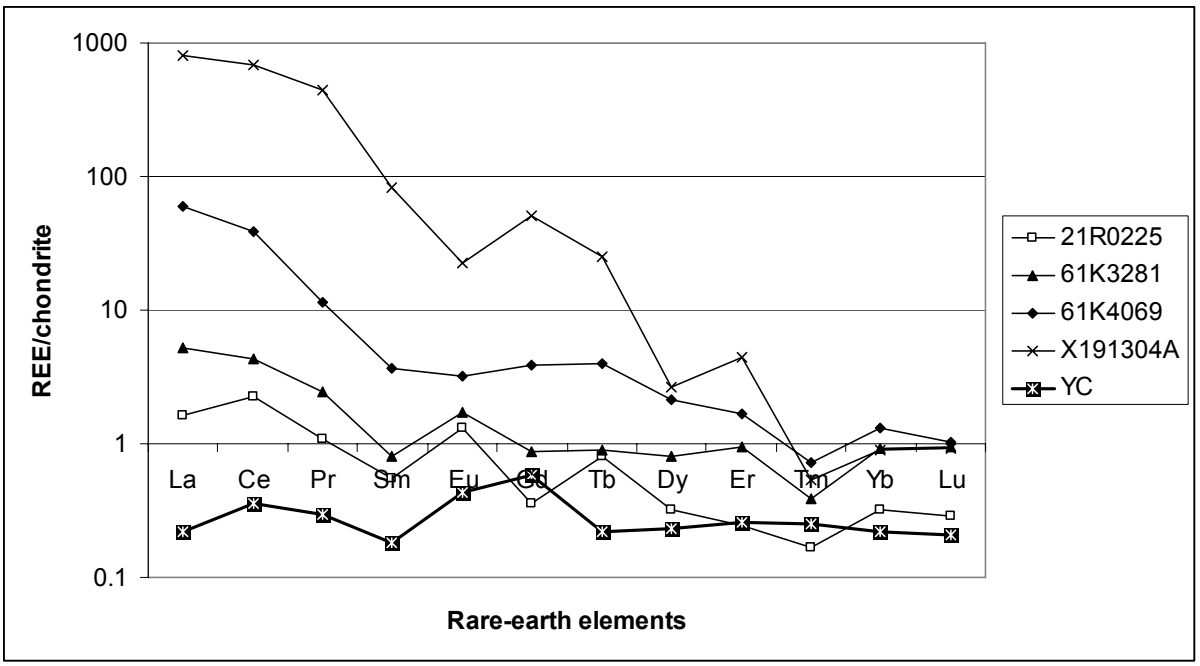

Figure 2: REE pattern of the yellow cake and the uranium ore samples originated from the different parts of Hungary (normalized values to chondrite)

Comparing the patterns obtained to published data [10], the origin of yellow cake sample is most probable sandstone. However, the production technology of the Hungarian yellow cake is a ground water purification procedure therefore the obtained REE pattern may change during the heap leaching. Nevertheless, it is known that the geological composition of the Mecsek Mountains is mainly sandstone.

It is important to note that uranium ore samples indicated in Figure 2 with 21R0225, 61K3281, 61K4069 and X191304A signs do not originate from the Mecsek region. The origin of these ores in order as mentioned above is: Lake Balaton, Bükk Mountains (61K3281, 61K4069) and Sopron (different parts of Hungary). The REE patterns of these ores show differences to the ones originate from the Mecsek Mountains. More similarity can be seen in Figure 1 among the REE pattern of uranium ore and yellow cake samples.

Studying the pattern of the different origin ores shown in Figure 2 the observed differences can be utilized for origin assessment of ore and yellow cake samples. The uranium ore sample e.g. signed with X191304A originating from Sopron can be excluded as the raw material of the uranium ore concentrate based on its Pb isotope ratio. This ratio ($^{208}Pb/^{204}Pb$: 104±2) indicates that it is probably a thorium ore which was proven by the measured high Th content (113±1 μg g^{-1} contrary to the average 0.5 – 1 μg g^{-1} in the other ore samples).

In addition, two figures below show the morphology and structure of the uranium ore concentrate. Figure 3 is an image taken by optical microscopy. Figure 4 is an image taken by SEM at ITU.

Figure 3: Optical microscopy photo of the Hungarian uranium ore concentrate taken at ITU

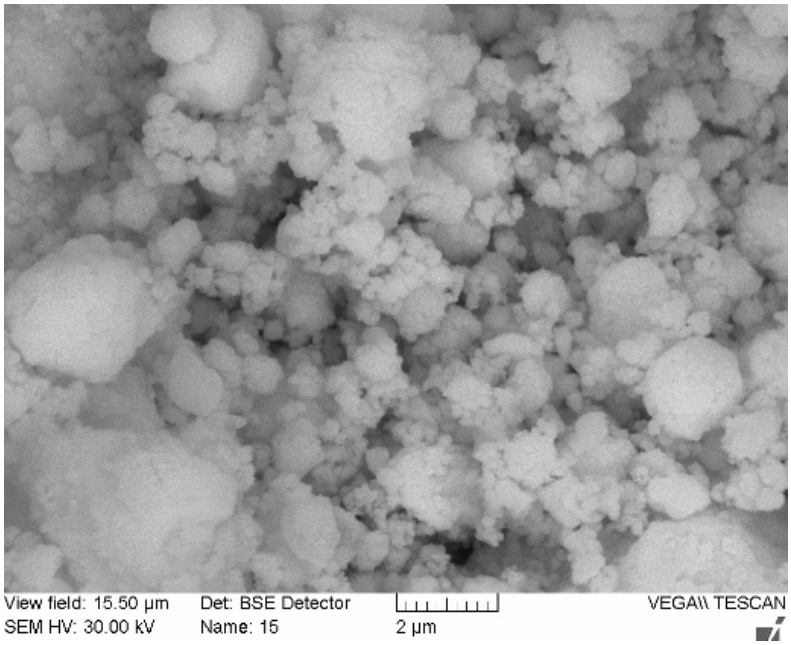

Figure 4: SEM image of the Hungarian uranium ore concentrate taken at ITU

The production date was determined at ITU as 11.4 ± 0.7 years (ref. date: 08/12/2010), corresponding to July 1999 ± 0.7 years. The reference value is: 10/1999.

The uranium ore concentrate was also analyzed by FTIR at ITU. The sample is $UO_4.2H_2O$, which is precipitated or dried at elevated temperature [11]. The major regions in the FTIR spectrum: two overlapping broad bands at 3470 and 3140 cm^{-1} due to the O-H stretching vibrations; often a sharp peak at 3330 cm^{-1} from the OOH stretching is also visible; a band with high intensity due to the vibrational bending mode of H_2O (δH_2O) at 1620 cm^{-1}; a strong band at 920 cm^{-1} is the asymmetric stretching frequency of the uranyl group; a band at around 490 cm^{-1} from the U-O stretching . The sample contains nitrate and smaller amount of carbonate, which are expected to derive from the precipitating liquor (in our case carbonate comes from the leaching solution, nitrate is an impurity from the ion exchange purification). The IR spectrum can be seen in Figure 4.

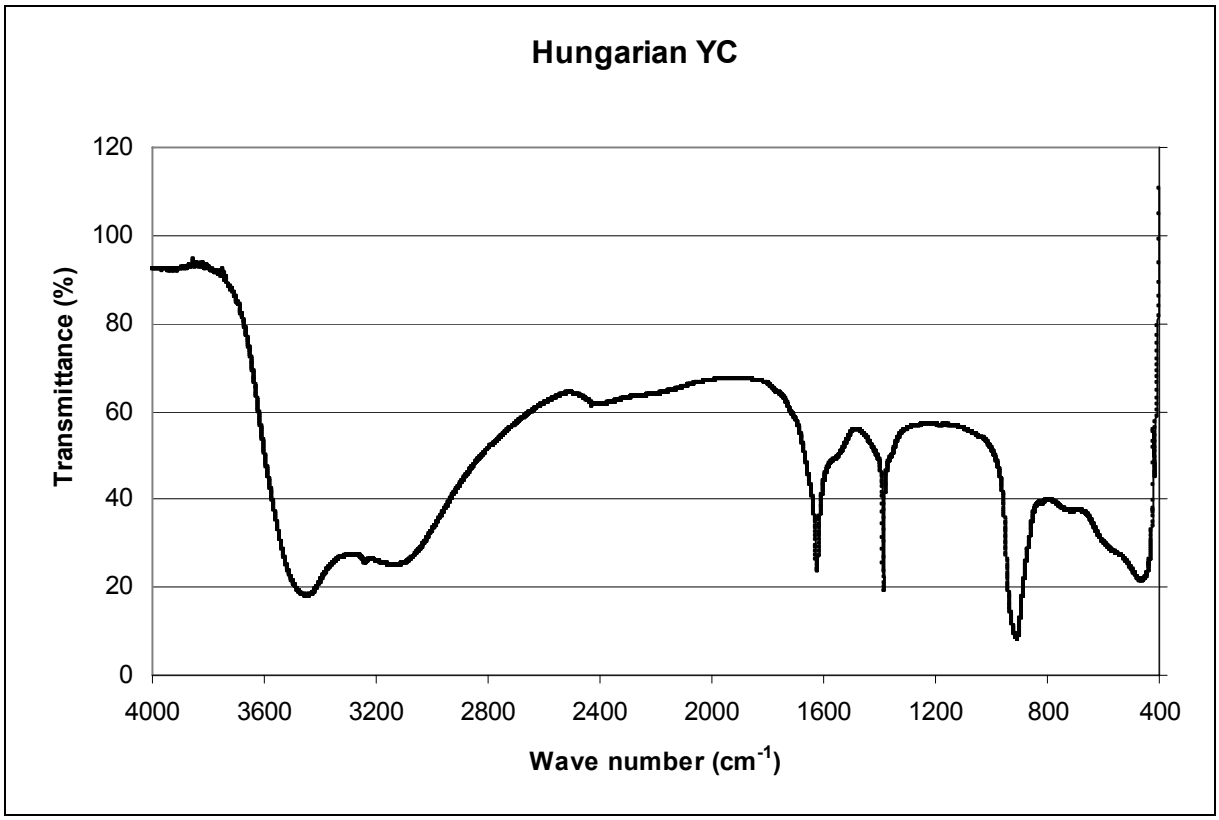

Figure 4: FTIR spectrum of Hungarian yellow cake sample

4. Conclusions

Uranium ore concentrate sample was analyzed at ITU and IoI in the frame of a collaboration agreement in nuclear forensics assistance. Results were compared and good agreement was found within the combined uncertainties. Some examples from analysis of uranium ore samples were also presented in the paper. Differences among the analytical results of ITU and IoI originated most likely from the inhomogeneity and differences in sample preparation method used for the uranium ore samples.

The joint analysis exercise demonstrates well two aspects, which have greatest importance in the field of nuclear forensics and, in the larger scale, in nuclear security. First, reasonably good analytical capability to investigate seized nuclear materials is required. Their usage can be often extended from basic analysis to specialized nuclear forensics analysis by training and such "intercomparison" exercises as illustrated here. Secondly, in order to receive nuclear forensics assistance (in the frame of real seized sample analysis), it is important to have a collaboration agreement in place that in case of an incident, the support, if needed, can be requested in a swift manner.

5. Acknowledgement

The authors wish to thank the co-workers of MecsekÖko Zrt., especially Dr. Zoltán Máthé and the Hungarian Atomic Energy Authority for the uranium ore and concentrate samples. Tamás Bíró's efforts, scientific advices and his help in the shipment of the samples were also highly appreciated.

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18 NDA II - Neutron data acquisition and analysis

Improvements in Dead-time Correction Using List-mode Neutron Counters

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Abstract:

Neutron multiplicity counting is a widely used technique in safeguards to determine the mass of fissile material. Unfortunately the multiplicity measurement is disturbed by the dead-time effect, which prevents pulses from being detected by the electronic chain on the same channel when they follow shortly in time after another pulse. This is particularly important since this loss due to dead-time especially affects neutrons originating from multiple-emission events, which in fact carry a lot of information about the sample and so should be measured. There exist a number of different dead-time correction methods. The most widely used (INCC) makes use of a semi-empirical correction for the single and double neutron emissions (Singles, Doubles) and another method (of Dytlewski) for Triples. They require the application of a pre-delay and of dedicated calibration measurements in order to determine the necessary parameters. Other correction methods have had little in-field use because of the complexity of their formulation.

In this paper we present a new approach for correction, based rather on mathematical-combinatorial theory than on heuristic formulas suggested by the physical principles. In contrast to other existing methods this correction is applied to the multiplicity distributions of measured "Reals plus Accidentals" and "Accidentals". Test results for this new method using simulations are presented as well as comparisons to other, well established methods for dead-time correction. This new technique requires the deployment of new, so-called multi-channel list mode counters.

This new approach will result in a number of advantages over the heuristic one: since the Singles, Doubles, Triples, etc. are calculated later from the multiplicity distribution, this method will be suitable to correct also for higher number of simultaneous neutron emissions than Triples (Quadruples, Quintuples). Since this method is based on the collected data itself, no prior calibration will be necessary. Furthermore use of a pre-delay may not be necessary, leading to an increased gate-fraction and – in principle – to increased quality of the results. Although this method is still under development, it has already demonstrated good results for low and medium count-rates.

Keywords: neutron, multiplicity counting; dead-time correction; list-mode counters

1. Introduction

A neutron multiplicity counter usually consists of a big tube of polyethylene moderator with a sample cavity in the middle. Gas proportional counters, often ^{3}He tubes, are embedded in the polyethylene moderator see Figure 1. If a neutron is emitted from the source to be measured, it has a high probability to collide with the hydrogen in the polyethylene. It loses energy until it reaches thermal equilibrium with the material. Then it diffuses around the moderator until it is lost or captured. A

fraction of these thermalised neutrons eventually hits a gas-atom from the embedded gas tubes and results in a digital pulse. During this process of collisions with other atoms the neutron loses its information about its point of origin and original direction. Hence the actual ^{3}He tube where it will be registered can be seen as sufficiently random and uncorrelated to other neutron events. This feature is the basis of the method presented here.

If a preamplifier receives a pulse, it processes it and sends out a digital pulse, see Figure 2. These digital pulses are subsequently collected by a counter, in this case a multi-channel device. Although the time of the pulse train is continuous, the counter needs to break it up into discrete time intervals, synchronized between the channels, which, in the following explanation are called “TICs.”

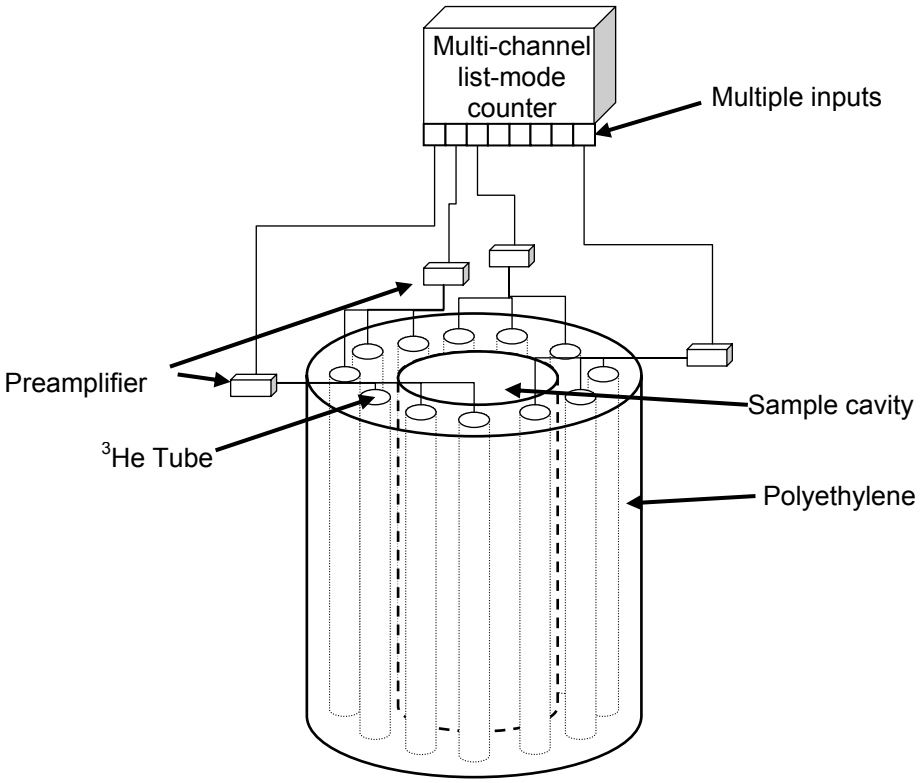

Figure 1: Multi-channel list-mode counter with detector

After the detection of a neutron, the ^{3}He tube and the connected preamplifier needs some time to recover (usually of the order of a microsecond), in which no further pulse can be received from this tube, even if another neutron is captured within this tube. This well known effect is called “dead time effect”. It is especially disturbing since during multiplicity counting one particularly wants to measure pulses that follow other ones after a short time. The “dead time” is the time within which counts may get lost due to this effect and is characteristic for the detector.

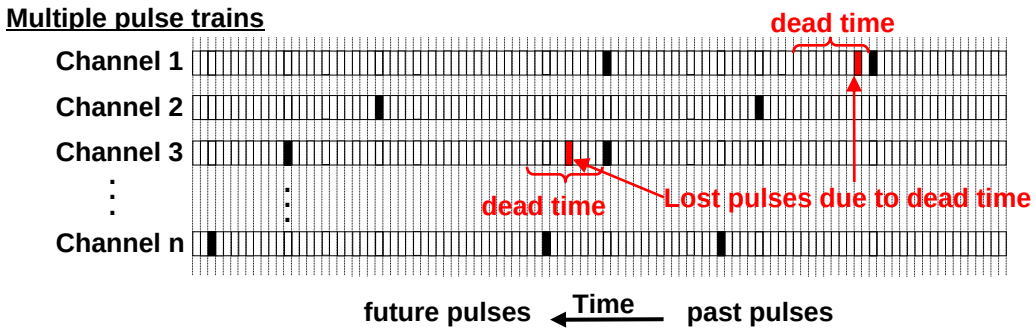

Figure 2: Pulse train from multi-channel list-mode counter

However, only pulses which come from the same preamplifier as a previous pulse are subject to loss within the dead time. Hence for multiple channel counters where the different channels are wired up to separate preamplifiers, no pulse can arrive at the same channel shortly after a pulse had been registered at this channel, whereas there is no limitation on pulses between different channels.

2. Principles of neutron analysis

The Rossi-Alpha distribution (see Figure 3) is obtained by "dropping" each pulse appearing after a certain timeframe of a leading pulse (called trigger) into a bin corresponding to its distance (in TICs) from the trigger pulse. This is repeated using each pulse of the pulse stream as the trigger pulse. The pulses in the respective bins are summed up with those already collected there. The counts that are collected in this way are called the Rossi-Alpha distribution. In contrast to the classical Rossi-Alpha distribution, where the process is restarted with the next pulse after the end of the gate (see Ensslin [1]), we do this for every pulse in order not to lose any information. Hence the timeframes from different trigger-pulses partially overlap. The Rossi-Alpha distribution, originally used for reactor noise analysis, is flat for a purely random source and is enhanced at early times close to the trigger when correlated events are present.

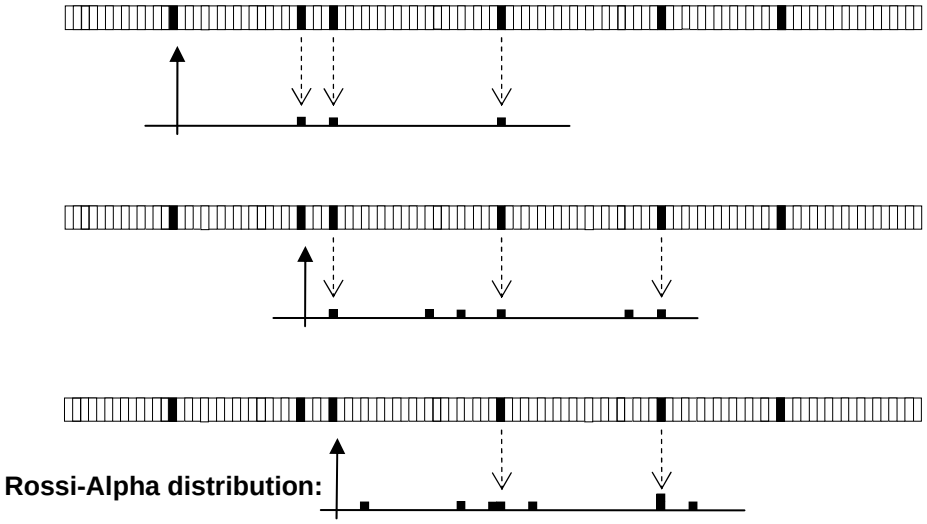

Figure 3: Building a Rossi-Alpha distribution

For neutron multiplicity counting the data analysis is different. One counts the number of pulses (called multiplicity) within a time window of certain length (called gate) opened after a leading pulse (called trigger). This process is applied for each pulse of a pulse stream used as trigger, the resulting statistics is called multiplicity distribution, see Figure 4.

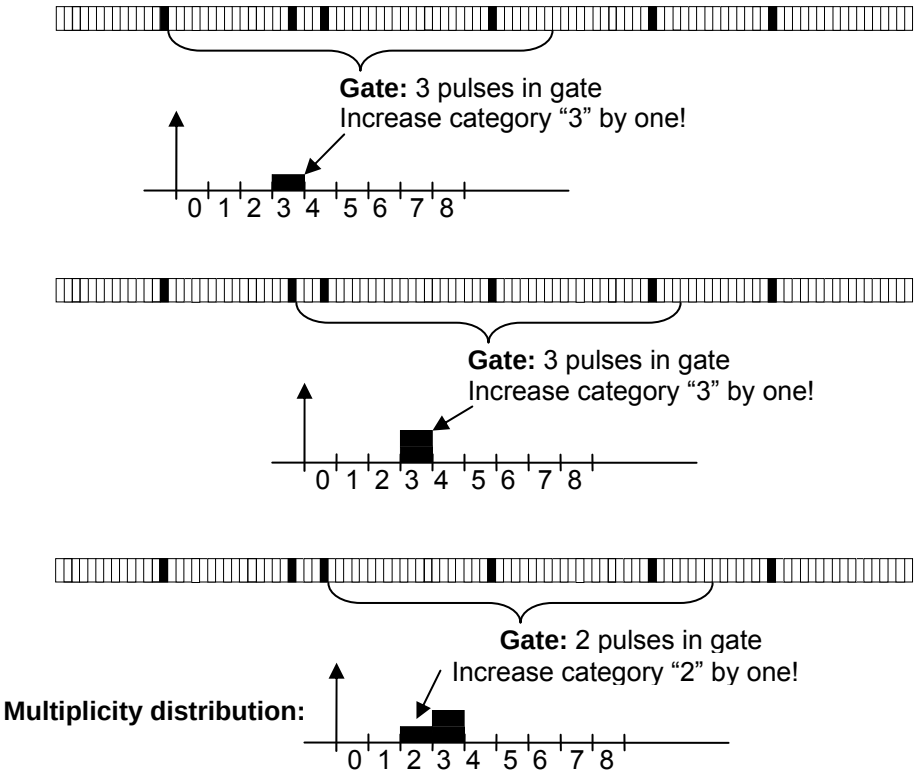

Figure 4: Building a multiplicity distribution

Usually there are two multiplicity distributions collected, one with the gate opened shortly after the trigger (called “Reals plus Accidentals”, or RA), another one measured by opening the gate after a long delay (1 to 4 msec.) after the trigger. After this long delay, which is many times the neutron lifetime within the detector, there is no remaining time-correlation between the pulse-distribution within the gate. Hence an accidental multiplicity is measured, the resulting distribution called “Accidentals”, or A. Nevertheless, both the RA and the A distribution suffer from dead-time losses.

The real single (*S*), double (*D*), triple (*T*) detection of pulses within the gate, with the contribution from accidentally multiple neutron detections removed, is calculated according to the following, well known combinatorial formulas:

$$S = \sum_{i=0}^{\max} RA_i \qquad \text{Eqn. 1}$$

$$D = \sum_{i=0}^{\max} i * RA_i - \sum_{i=0}^{\max} i * A_i \qquad \text{Eqn. 2}$$

$$T = \sum_{i=2}^{\max} \frac{i*(i-1)}{2} RA_i - \sum_{i=2}^{\max} \frac{i*(i-1)}{2} A_i - \frac{\sum_{i=1}^{\max} i * A_i}{\sum_{i=0}^{\max} A_i} * \left(\sum_{i=1}^{\max} i * RA_i - \sum_{i=1}^{\max} i * A_i \right) \qquad \text{Eqn. 3}$$

Although some theoretical work has been done by Hage and Cifarelli [2], for correcting these values, semi-heuristic approaches are used up to now, e.g. the (A,B)-correction (see Swansen [3]):

$$S^{cor} = S * e^{(a+bS)*S/4} \quad \text{Eqn. 4}$$

$$D^{cor} = D * e^{(a+bS)*S} \quad \text{Eqn. 5}$$

In the above formulas *S*, *D* denote the rate of singles and doubles where S^{cor} and D^{cor} denote the corrected singles and doubles rate respectively. The most commonly used Triples correction is given by Dytlewski [4] and is explained for practical use by Harker and Krick in [5].

All these methods require dedicated calibration measurements for each detector in order to adjust parameters (*A* and *B* in case of the (A,B)-correction and the 'multiplicity deadtime' δ_{mult} and C in the case of Triples) used in these heuristic formulas.

3. Direct correction of multiplicity distribution

Due to the increased number of channels, modern multi-channel list mode counters provide increased possibilities for neutron data analysis. In addition to the classical multiplicity distribution, further statistics on the arrival time of neutrons, channel number, cross- and auto-correlation, and triggered multiplicity can be collected. One new analysis is the use of the observed differences in coincidence rates between different channels to perform a dead-time correction to the complete multiplicity distribution.

3.1. Correction for lost triggers

In Figure 5 one can see a Rossi-Alpha distribution from experimental data collected from a Cf source with 6.14×10^4 neutrons/sec emission rate. An epithermal neutron counter (ENMC) from Los Alamos [7] comprising 27 preamplifiers and hence 27 channels for counting was used. One TIC corresponds to 0.1 μs. The blue part shows the neutrons captured on a different channel (and pre-amplifier) from the trigger ("*cross-correlated*"), the red part on top shows the neutrons captured on the same channel ("*auto-correlated*"). One can clearly see that, due to the dead-time effect, the first few fractions of a microsecond after the trigger there are no auto-correlated pulses arriving.

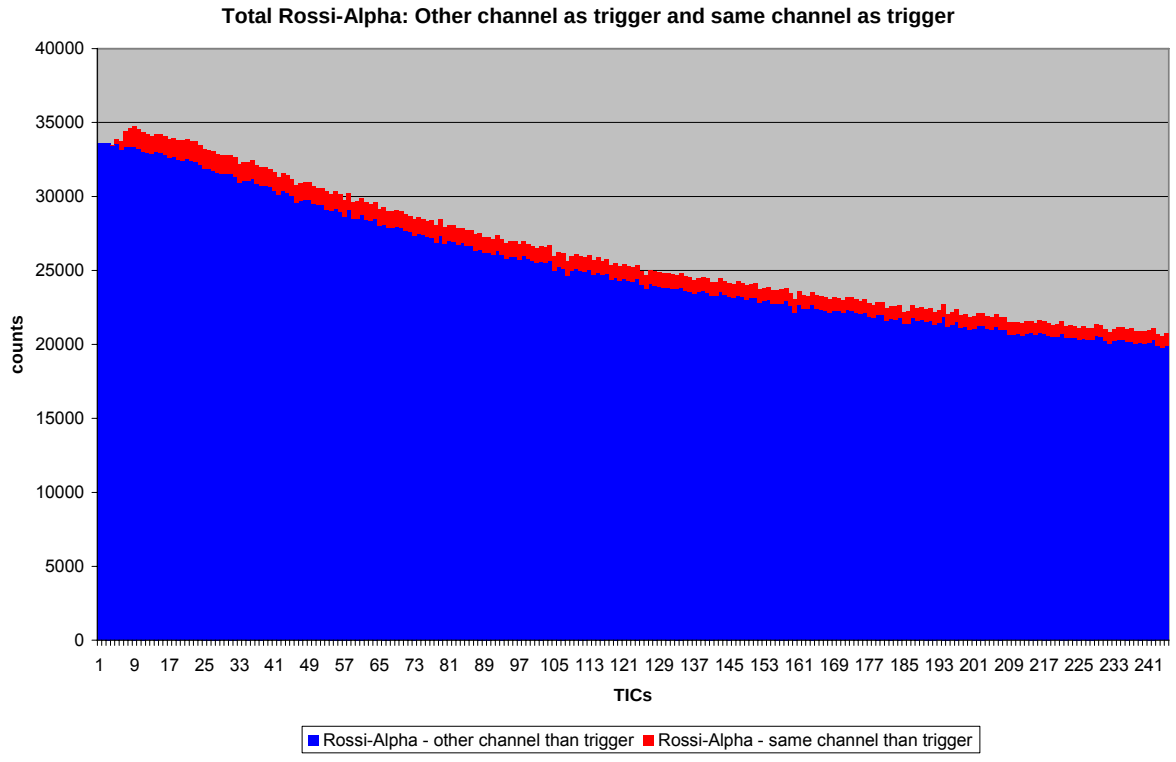

Figure 5: Rossi-Alpha distribution of a Cf source, displaying cross- and auto-correlated pulses

The pulses missing due this dead-time effect may be estimated by scaling up the cross-correlated pulses (blue part seen in Figure 5) to the overall count-rate and then subtracting the overall Rossi-Alpha distribution from it. The result can be seen in Figure 6. Since every pulse would become trigger if it had not been lost, this corresponds to the number of lost triggers.

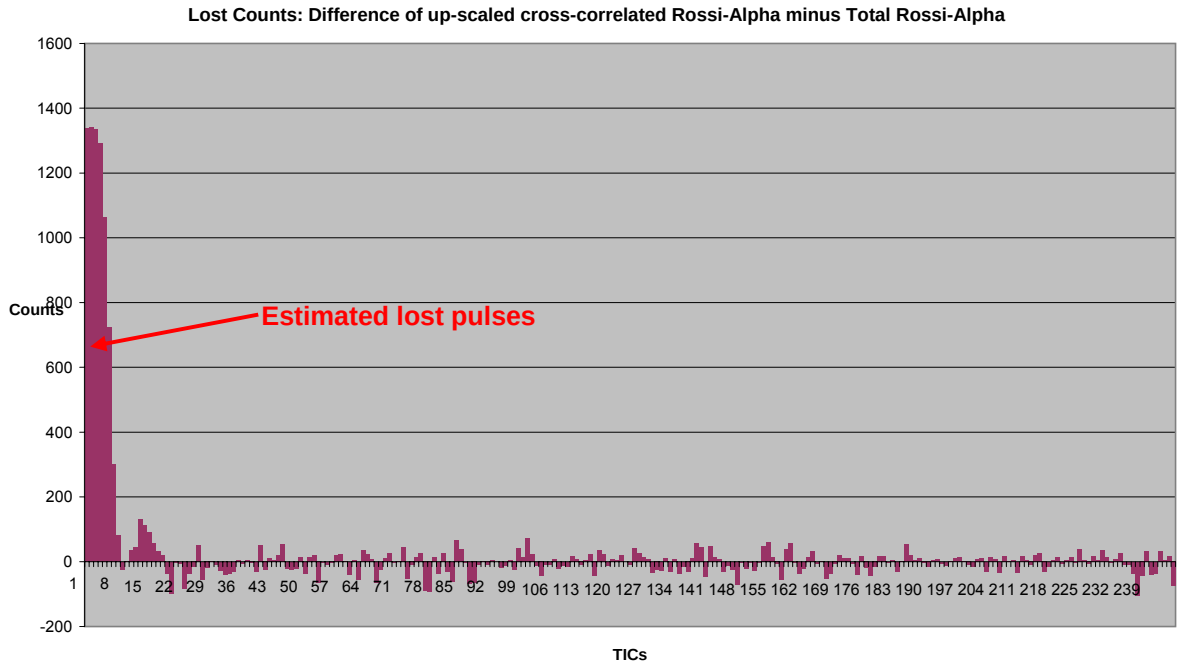

Figure 6: Estimated lost pulses (and triggers) of Cf source

When building the Rossi-Alpha distribution one can also, in parallel, store the multiplicity value (the number of pulses in the multiplicity gate) for each trigger together with the distances to its predecessors. This results in a time–distribution of triggers and their multiplicities. Figure 7 gives this time/multiplicity-distribution of triggers (in % of the total within that time-slot, up to a multiplicity of 15) with respect to the distance of leading pulses for the same source as before. One can see that the lower multiplicities (0 and 1) decrease when the trigger is closer to preceding pulses and therefore higher multiplicities portions increase. Since counts (and therefore triggers) are lost only when the time between pulses is short, this effect must be taken into account for any correction for lost triggers due to dead-time.

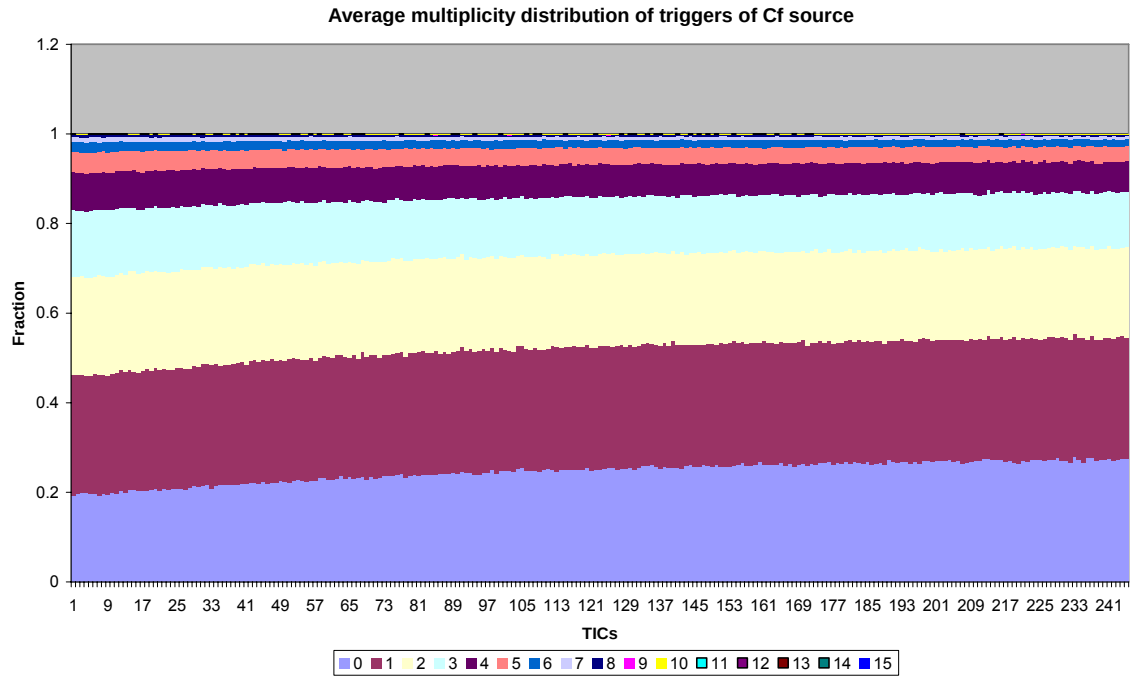

Figure 7: Multiplicity distribution of triggers with respect to distance from preceding pulses

The correction for lost triggers is then done in the following way. For each time-slot the estimated lost pulses as seen in Figure 6 is multiplied by the multiplicity distribution of the same time-slot found in Figure 7. This is summed up until there is no significant difference, which then gives a multiplicity vector $\overrightarrow{LT}$ for the lost triggers. Assuming that the lost triggers behave in the same way than those measured, one can correct for lost triggers by adding the lost trigger multiplicity distribution $\overrightarrow{LT}$ to the observed multiplicity distribution.

$$\overrightarrow{RA^{LT-corr}} = \overrightarrow{RA} + \overrightarrow{LT} \qquad \text{Eqn. 6}$$

More details on data collection and the background of the subsequent calculations can be found in Holzleitner and Swinhoe [8].

3.2. Correction for lost pulses inside a gate

In a similar way, by applying this procedure above to all pulses inside a multiplicity gate and by taking the multiplicity of the gate inside which these pulses occur as characteristic, one can estimate missing gates of a certain multiplicity category (see Holzleitner and Swinhoe [8] for details). This results in an "upgrade vector" $\overrightarrow{UG}$. However, one cannot simply add this vector to the multiplicity distribution as done for the lost triggers, since a multiplicity value was already obtained, possibly in a wrong multiplicity category if it was affected by deadtime. By assuming that the count-rate is low enough to have only one pulse lost inside a gate, one can correct as seen in Eqn. 7. The subtraction of the UG_{m+1} one multiplicity higher is because the higher multiplicity value had already been counted, just in a multiplicity category one lower than it should have been due to the missing pulse inside the gate.

$$\overrightarrow{RA^{corr}} = (RA_m^{corr})_{m=0,\ldots} = (RA_m + LT_m + UG_m - UG_{m+1})_{m=0,\ldots} \qquad \text{Eqn. 7}$$

Here, the assumption that only one pulse had been lost inside a gate had been made. This holds only for low to medium count-rates and/or short gates. Despite this restriction, this method provides good results for low and medium count-rates as it will be demonstrated in the following section. Loss of more than one pulse in a gate will be subject to further analyses in future.

4. Simulation and comparison

Since a neutron lost due to dead time is not observed, one has to use simulations and comparisons to existing correction methods in order to determine the usefulness of the present method. In the following a simulation is presented, later we use some measurement results to compare the presented correction to the (A,B) correction (see Swansen [3]) and Triples correction (see Dytlewski [4] or Harker and Krick in [5])

4.1. Simulation

We used a simulation of 143630 counts/sec, 100 sec count time and an assumed time resolution of 1 TIC = 100 ns. Decays that are exponentially distributed in time (Poisson distribution) are simulated, each decay emitting between 1 and 7 neutrons, the neutron distribution can be seen in Figure 8. After 1 μs the neutron is assumed to be thermalised, then it is assumed to be registered after an exponential time-distribution by one of 32 channels, equally distributed between the channels. 100% efficiency of the simulated detector is assumed. This results in an original pulse train unaffected by dead time.

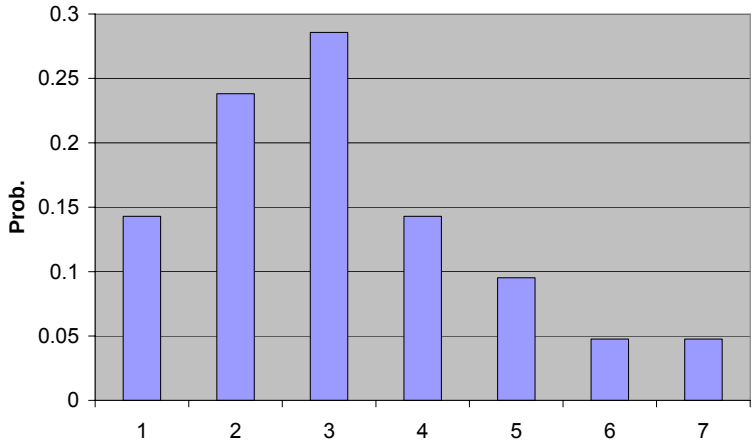

Figure 8: Probability of number of neutrons emitted per simulated decay

Dead time is applied to this pulse train by using a deadtime distribution rather than a fixed deadtime value. This is done by deleting any pulse appearing 1-5 TICs after a leading pulse on the same channel, 6 TICs after a leading pulse it is deleted with 75% probability, 7 TICs later with 50% and 8 TICs later with 25% probability. Any other pulse remains unchanged. This gives the “dead-time affected” pulse train.

Both pulse trains can now be analysed according to their multiplicity distribution, where a gate of width 24.5 μs was used. No pre-delay for the RA gate in the classical sense was used and a long delay for the A gate of 1000 μs was used. The difference between the dead-time-affected and the non dead-time-affected multiplicity distributions, both for RA and A, can be seen as the dark blue and magenta curves in Figure 9. After applying the correction described here the differences were taken again both for RA and A, and can be seen by the yellow and light blue curves in Figure 9. As one can see, these curves lie around the zero line, which means that the difference between the corrected and the original multiplicity distribution (unaffected by deadtime) is around zero with some variance. The correction for A results in a higher variance, the reason is not yet known.

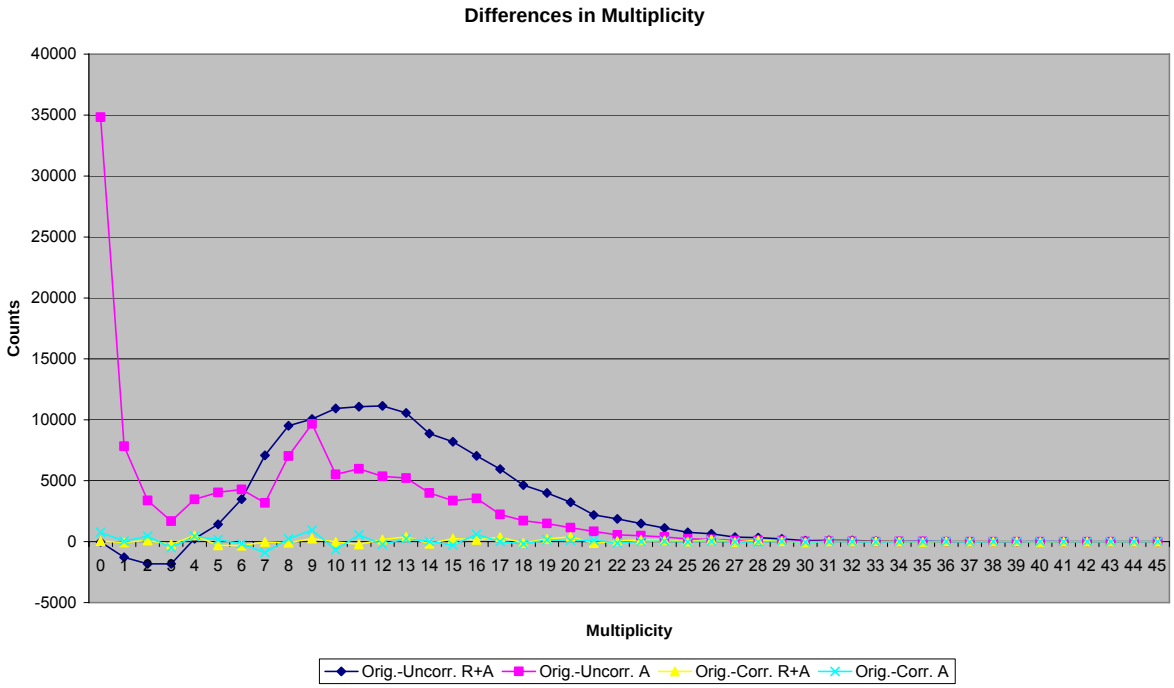

Figure 9: Differences of original minus dead-timed and original minus corrected multiplicity distributions

4.2. Comparison with other correction methods

Here we want to give a brief comparison of this new correction method with other correction methods. Especially we compare results from real measurements with the (A,B) correction by Swansen [3] for the Singles and Doubles rates and the Triples rate correction by Dytlewski [4], both described in Harker and Krick [5].

Again the epithermal neutron counter (ENMC) from Los Alamos [7] comprising 27 preamplifiers and hence 27 channels for counting was used. This multiplicity counter uses 10-atm ^{3}He tubes and has an efficiency of 65% and a 22-µs die-away time. For all evaluations a gate with of 24.0 µs, a predelay of 1.5 µs, and a long delay of 4096 µs was used. The dead-time parameters for this detector can be seen in Table 1.

(A,B) correction for Singles / Doubles	A	0.0954 µs
	B	0.0289 µs^2
Triples correction of Dytlewski:	δ_{mult}	0.0368 µs
	C	0

Table 1: Dead-time parameters for ENMC neutron detector of Los Alamos National Laboratory

Source	Measured Singles (counts/sec)	(AB) method for Singles		Method described here:	
		Corrected Singles	Diff from uncorrected:	Corrected Singles	Diff from uncorrected:
AmLi ~10^5 neutrons / sec. emission, ~400 sec meas.-time	63877.41	63976.69	+99.28	63976.51	+99.10
AmLi ~1.75×10^5 neutrons / sec. emission,, ~25 sec. meas.-time	113566.05	113884.68	+318.63	113975.19	+409.14

AmLi ~3.75×10^5 neutrons / sec. emission,, ~10 sec. meas.-time	243415.88	244937.98	+1522.10	245364.81	+1948.93

Table 2: Correction of AmLi sources of various strength, using different measurement times.

Corrections of Cf source with 6.14×10^4 neutrons/sec.	Measured Rate	(AB) method for Singles / Doubles or Dytlewski method for Triples respectively:		Method described here:	
		Corrected Rate	Diff. from uncorrected:	Corrected Rate	Diff. from uncorrected:
Singles rate:	38889.24	38925.34	+36.1	38961.95	+72.71
Doubles rate:	24514.47	24606.40	+91.93	24642.84	+128.37
Triples rate:	8639.2	8876.35	+237.15	8808.64	+169.44

Table 3: Correction of a Cf source with 6.14×10^4 neutrons/sec emission rate, 400 sec. measurement time.

Corrections of Cf source with ~4.9×10^5 neutrons/sec.	Measured Rate	(AB) method for Singles / Doubles or Dytlewski method for Triples respectively:		Method described here:	
		Corrected Rate	Diff. from uncorrected:	Corrected Rate	Diff. from uncorrected:
Singles rate:	315707.29	318322.56	+2615.27	318946.53	+3239.24
Doubles rate:	193907.18	200412.67	+6505.49	201291.54	+7384.36
Triples rate:	55420.51	67867.50	+12446.99	65358.17	+9937.66

Table 4: Correction of a Cf source with ~4.9×10^5 neutrons/sec emission rate, ~10 sec. measurement time.

Tables 2-4 show the results of the traditional correction method with those of the new correction proposed here. They show that for this data the two correction methods give similar results. This is satisfying and to be expected as the existing deadtime corrections have performed well for in-field measurements at these counting rates. From this experimental data alone it is not possible to decide which is the better method, but the current method can also give corrected values for Quads etc.

5. Conclusion

This new method of deadtime correction presented here provides several advantages: The two main advantages are clearly that, since the Singles, Doubles, Triples, Quads, etc. are calculated later from the multiplicity distribution, this method will be suitable to correct for any such multiplicity. Secondly the correction is based on the collected data and so no previous dead-time measurement for calibration is necessary. Furthermore this method does not use a classical pre-delay, hence the increased gate-fraction leads, in principle, to the increased quality of the results. This correction technique could be implemented in the hardware of the instrument itself.

A comparison with simulation has shown that the multiplicity distribution corrected for deadtime by this method is very similar to the original unperturbed distribution. Also the magnitude of the correction for Singles, Doubles and Triples is similar to existing deadtime correction methods for some Cf and AmLi measured data.

The accuracy of the correction depends on the count-rate per channel and can certainly be improved for higher count-rates using a more sophisticated evaluation. It has been found that good results can

be provided for a count-rate of about 150 000 counts/second using 32 channels, provided that the count-rates per channel are about equal.

6. Acknowledgements

This work was done within the framework of the EURATOM - US/DOE agreement. The authors would like to thank all persons who contributed useful discussions and comments to this topic,

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Extension of ESARDA NDA Multiplicity Benchmark Simulations to Validate Dead Time Correction Algorithms

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Abstract:

Dead time correction algorithms are being evaluated at Los Alamos National Laboratory (LANL) as part of a strategic R&D program to advance correlated neutron analysis. Such algorithms have an immediate impact on currently and soon to be deployed neutron detection systems. The Monte Carlo N-Particle eXtended radiation transport code (MCNPX) can be used to generate a pulse train file that can be post-processed by bespoke analysis software that includes the simulation of dead time effects. This simulation methodology is known as Monte Carlo pulse train generation. Further analysis of the simulated pulse trains with multiplicity shift register (MSR) algorithms can be used to study the dead time perturbed Singles, Doubles and Triples counting rates. MSR analysis is being included in the scope of work since many currently deployed counters cannot benefit from pulse trains acquired using list mode data acquisition, because they are constrained by hardware.

In February 2006 the ESARDA non-destructive assay (NDA) working group published multiplicity benchmark (phases I and II) results in the ESARDA bulletin number 34. This benchmark compared the codes and analysis algorithms used for the simulation of neutron multiplicity counters. The basis for comparison was a series of neutron pulse trains generated by LANL using MCNPX. This paper will present the Monte Carlo pulse train methodology developed for this benchmark exercise and the simulation of dead time effects. The original benchmark simulations have been re-visited to explore the effects of dead time and extended in an attempt to validate dead time correction algorithms. The simulation work in this study accounted for the total dead time in the detection system, based on an extending (or paralyzable) model.

Keywords: NDA; Monte Carlo; dead time; neutron multiplicity; nuclear safeguards

1. Introduction

Temporally correlated neutron emission, from both spontaneous and induced fission, provides a unique signature for the detection of special nuclear material (SNM). Passive neutron multiplicity counting based on multiplicity shift register (MSR) electronics uses signal triggered and (what amounts to) randomly triggered gate inspection to record the number of occurrences of multiplicity i to generate MSR histograms and thus derive correlated neutron event rates. This technique is commonly used in international safeguards measurements for the non-destructive assay of plutonium (Pu) mass.

Denoting the signal triggered and randomly triggered histograms by N_i and B_i and the assay time by t the observed or measured Singles, S_m, Doubles, D_m and Triples, T_m, rates may be expressed by Equations 1, 2 and 4, respectively.

$$S_m = \frac{1}{t} \cdot \sum_{i=0}^{\infty} N_i = \frac{1}{t} \cdot \sum_{i=0}^{\infty} B_i \tag{1}$$

The Singles rate is also referred to as the Totals rate or Gross neutron rate and importantly can also be thought of as the MSR trigger rate, which is the rate of inspection interval or coincidence gate openings.

$$D_m = \frac{1}{t} \cdot \sum_{i=1}^{\infty} i \cdot \left(N_i - B_i\right) \tag{2}$$

The expression for the Doubles is formed by the difference between the genuine (or Real) plus chance (or Accidental) pairs tallied with the trigger and the Accidentals. It therefore follows that the Accidentals coincidence (or pairs) rate for Doubles counting, is derived from the random triggered histogram, and takes the form given by Equation 3.

$$A_m = \frac{1}{t} \cdot \sum_{i=1}^{\infty} i.B_i \tag{3}$$

$$T_m = \frac{1}{t} \cdot \sum_{i=2}^{\infty} \frac{i \cdot \left(i-1\right)}{2} \cdot \left(N_i - B_i\right) - A_m \cdot \frac{D_m}{S_m} \tag{4}$$

The last term in the Triples expression reduces to the product of the Singles, Doubles and gate width; thus may be thought of as Doubles randomly promoted to Triples.

1.1. Dead time

Currently deployed neutron multiplicity counting systems utilize ^{3}He proportional counters for neutron detection, served by dedicated electronics which include amplifier-discriminator boards (~ 3-10 tubes per board) followed by an OR gate or derandomizer circuit. Only signals which exceed the discriminator threshold are counted (generate a digital pulse). The next signal cannot be counted reliably until the trace has returned to a value below the threshold, otherwise pulse pile-up is encountered. This is considered to be one of the main sources of dead time in a neutron multiplicity counting system. The action of the discriminator explains one aspect of counting losses and provides justification for the use of an extending dead time model in this work. In simulation we are able to recreate this ideal model, however real life is more complex. In reality, the system dead time may resemble a combination of extending and non-extending dead times, or indeed some other manifestation.

1.2. Dead time mitigation by design

Dead time effects may (to some extent) be mitigated by design; by distributing the counts between many ^{3}He proportional counters with a single amplifier-discriminator board per counter. However, this is a costly approach, which extends the current methodology to higher count rates but does not solve the problem. It would be especially costly to retrofit new electronics to all the neutron multiplicity counters deployed in the field. Mitigation by design also has the drawback that the system dead time has a fundamental lower limit dictated by the collection time of the electrons from gas ionization and amplification. Given these limitations, there is still a need for accurate dead time correction methods during analysis of the measured neutron multiplicity data.

1.3. Review of traditional Singles rate dead time correction

Dead time correction algorithms, or correction factors, are applied to measured neutron counting rates to correct for counting losses and thus derive estimates of the true counting rates i.e. the counting rates expected in the absence of dead time. As will be described, correction factors have traditionally been applied solely to the measured neutron counting rates (Singles, Doubles and Triples) using formalisms based on the assumptions of an extending (paralyzable) dead time and Poisson source. In this paper, we consider the correction of the MSR histograms with the advantage that the histograms are assay item specific and corrections are therefore not reliant on the same assumptions.

The Singles rate dead time correction factor CF_S is the ratio S_c/S_m of the dead time corrected Singles rate to the measured (uncorrected) Singles rate. This correction factor should be unity in the absence of dead time. The traditional Singles rate dead time correction factors that we will consider in this paper are herein known as (1) the Poisson Singles rate dead time correction factor, (2) the Empirical Singles rate dead time correction factor, and (3) the Dytlewski Trigger Singles rate dead time correction factor. (4) A fourth alternative histogram-dependent Singles rate dead time correction factor will also be proposed and referred to as the Croft-Dytlewski Singles rate dead time correction factor.

1.3.1. Poisson Singles rate dead time correction factor

The exact Singles rate dead time correction factor for a random (Poisson) neutron source subjected to extending dead time would be of the form given by Equation 5.

$$CF_S = e^{d \cdot S_c} \tag{5}$$

where d is the multiplicity dead time parameter (value of the fixed extending dead time) [1]. AmLi sources approximate the Poisson condition, whereas ^{252}Cf and Pu items have correlated neutron pulse trains.

1.3.2. Empirical Singles rate dead time correction factor

In traditional neutron coincidence counting (Singles, Doubles), the extending dead time correction factor is applied as an empirical two-parameter approximation, given by Equation 6, which is the Doubles dead time correction factor raised to the fourth root in the exponent.

$$CF_S \approx e^{\frac{1}{4}\delta \cdot S_m} = e^{\frac{1}{4}\cdot(A+B\cdot S_m)\cdot S_m} \tag{6}$$

where δ is distinct from d and is referred to as the effective Doubles dead time parameter. For equality with the Poisson model at low rates, we can define A and B in terms of d (although normally A and B are chosen as free parameters).

$$A = 4 \cdot d, \text{and } B = \frac{A^2}{4} \quad (7), (8)$$

Through experience [2], the inclusion of a higher order term in the exponent is beneficial. Currently, the safeguards software INCC makes use of the semi-empirical correction for the Singles and Doubles rates, and the Dytlewski [3] method for the Triples rate. In addition, a consistent dead time treatment could also be developed for both neutron coincidence and multiplicity counting (Triples).

1.3.3. Dytlewski Trigger Singles rate dead time correction factor

Dytlewski [3] proposes a Singles rate dead time correction factor, based on the correction for the loss of the trigger event rate due to dead time i.e. the lost number of gate inspection intervals. This correction factor is given by Equation 9.

$$CF_S \sim e^{d \cdot S_m} \quad (9)$$

where d is the system dead time parameter. The approximation is expected to be suitable at low rates $(d \cdot S_c << 1)$ and when the pulse train is not highly correlated. The level of correlation on the neutron pulse train can be formally quantified in terms of the ratio $(D_c / f_d) / S_c$, where f_d is the Doubles gate utilization factor [4]). D_c / f_d is the true (dead time corrected) Doubles event rate on the pulse train with 'perfect' gating so that no pairs are missed due to the pre-delay and gate width being of finite duration.

1.4. The need for improved dead time correction algorithms

As described, traditional dead time correction factors for neutron multiplicity counting are based on an extending (paralyzable) system dead time. Both the Singles and Doubles rate dead time correction factors are approximated by an exponential dependence similar to those for a pure random (Poisson) source. However, the pulse trains are not random for fission sources. There is a need to develop alternative dead time correction factors for the Singles, Doubles and Triples rates derived from the MSR histograms, to extend their application to correlated neutron sources. There is also a need to extend the application to higher order rates e.g. quads (quadruple correlations) etc. to enable a greater number of reliable correlated neutron rates to be obtained in order to reduce dependence upon calibration. Typically neutron multiplicity is used when the detection efficiency is known via the measurement of a number of calibration standards, but this requirement could perhaps be eliminated for complex items.

Evolving counter designs and safeguards applications also dictate a need for more rigorous analysis methods. Since the development of the first neutron multiplicity counters, detector designs have evolved to produce significant increases in efficiency and reductions in effective die-away time. In addition, the range of items being measured has expanded so that large masses of impure items in multiplying form are now routinely encountered. Shielded items may also pose a challenge. As a consequence of these changes, a greater concentration of correlated events appear on the neutron pulse train and management for the correction of dead time losses is being re-visited with a renewed focus.

1.5. Our Research Approach

Here, we extract a new alternative histogram-dependent Singles rate dead time correction factor and present this theoretical approach. The performance of this alternative correction factor is then compared to the three traditional Singles rate correction methods using the simulated neutron pulse trains created for the ESARDA NDA benchmark, perturbed for dead time in post-processing software. Conclusions are drawn as a first step towards developing and implementing advanced dead time correction algorithms. Conclusions regarding the simulation method can also be drawn and will be published elsewhere.

2. Croft-Dytlewski alternative histogram-dependent Singles rate dead time correction

Dytlewski [3] derives expressions for the dead time corrected Singles, Doubles and Triples counting rates, which themselves are derived from the MSR histograms. It can therefore be found that implicit within the Dytlewski dead time correction factors is an alternative form of the Singles rate dead time correction factor, based on the correction of the MSR histogram. The alternative form is beneficial because it is assay item specific in that it derives from the histogram. By identifying this alternative form, we aim to produce a set of self-consistent dead time correction factors so that dead time treatment for Singles (and in some implementations, Doubles) can be made within the same set of assumptions, model and calibration.

The Singles neutron counting rate can also be thought of as the MSR trigger rate. The trigger rate is the number of events that trigger the MSR coincidence gate structure to open and count correlated neutron events, which are then tallied as the MSR histograms. Since the Singles counting rate is perturbed due to counting (or dead time) losses, the number of trigger events is therefore reduced. The new alternative form of the Singles rate dead time correction factor compensates for the impact of counting losses on the trigger rate i.e. it compensates for the gates that did not open and thus did not contribute to the MSR histogram. This alternative form is given by Equation 10.

$$CF_s = \frac{S_c}{S_m} = \frac{\left(\sum_{i=1}^{\infty} \alpha_i \cdot B_i \Big/ \sum_{i=0}^{\infty} B_i\right)}{S_m \cdot T_g} \tag{10}$$

where T_g is the width of the coincidence gate (duration of the inspection interval). The ratio $B_i \Big/ \sum_{i=0}^{\infty} B_i$ is the normalized randomly triggered inspection interval multiplicity histogram. B_i is the number of events recorded in the randomly triggered (also called the Accidental or A-) histogram of multiplicity order i, t is the assay duration, and α_i are the dead time correction functions defined by Dytlewski [3] in terms of the extending dead time parameter d and the order of the histogram bin i. This form is inspired by the multiplicity dead time treatment introduced by Dytlewski [3] and implemented by Dytlewski, *et al.* [5] for Triples. Equation 10 does not collapse to Equation 5 for a Poisson source, but is a close approximation.

3. Simulation method

Three benchmark exercises have been carried out by the ESARDA Working Group on techniques and standards for Non-Destructive Analysis (NDA-WG) since 2003 in order to assess the capabilities of Monte Carlo modeling to reproduce experimental data and the capabilities of analyzing pulse trains. The ESARDA multiplicity benchmark is one such exercise, carried out to compare the different algorithms and codes used in the simulation of neutron multiplicity counters. The published ESARDA multiplicity benchmark exercise modeling data is used here to quantitatively test and compare the performance of four dead time correction schemes using simulated data that conforms to the constant extending dead time assumption.

The counter used to make measurements for the ESARDA multiplicity benchmark, and also used as the basis for the simulations, was the Active Well Coincidence Counter (AWCC) [6] in the fast configuration (Cd liner inside the cavity) in the passive mode (with both discs removed). This is a widely deployed and well known safeguards counter. This active well comprises a cavity height of 35 cm and utilizes 42 ^{3}He tubes for neutron detection. The performance characteristics of the AWCC include an efficiency of 28 % in the fast mode, and a die-away time of 50 microseconds [7]. An MCNPX model of the AWCC was produced by JRC Ispra.

3.1. Pulse train generation

As part of this benchmark exercise, Swinhoe [8] simulated thirteen neutron pulse trains, to represent a range of assay items and counting rates. Table 1 shows the cases that were simulated.

Case	Item
Case 1a	Random neutron source (AmLi) 10 kcps
Case 1b	Random neutron source (AmLi) 100 kcps
Case 1c	Random neutron source (AmLi) 1000 kcps
Case 2a	Spontaneous fission source (252Cf) 10 kcps
Case 2b	Spontaneous fission source (252Cf) 100 kcps
Case 2c	Spontaneous fission source (252Cf) 1000 kcps
Case 3a	Pu metal 10 g (90 % 239Pu)
Case 3b	Pu metal 1000 g (90 % 239Pu)
Case 4a	Pu oxide 10 g (reactor grade)
Case 4b	Pu oxide 1000 g (reactor grade)
Case 5a	Pu oxide 10 g (reactor grade) with AmLi source to give α = 10 (α = ratio of (α,n)/spontaneous fission neutrons)
Case 5b	Pu oxide 10 g (reactor grade) with AmLi source to give α = 20
Case 5c	Pu oxide 10 g (reactor grade) with AmLi source to give α = 100

Table 1: Simulated assay items

3.2. Dead time perturbation of pulse trains

The ESARDA NDA-WG did not extensively consider the simulation of dead time effects. Here, an extending dead time of fixed value was applied to each neutron event in the pulse train and the resulting dead time perturbed train was recorded and output to file. Several values of dead time were initially simulated, but the dead time parameter value of 180 ns closely represents the overall dead time of the AWCC (note that each preamplifier has a greater dead time). Dead time parameter values a factor of two above and below this value were also used for the final performance comparison.

3.3. Software MSR

An in-house software MSR known as VB-Tap6 was used to mimic the action of the MSR and apply dead time to the pulse train. This took the form of a fixed (rectangular) extending dead time after each pulse. The code was used to calculate the measured count rates for both the original pulse trains (without dead time losses) and the resulting dead time perturbed pulse trains.

4. Results

4.1 Effects of dead time on the MSR histograms

The effects of dead time on the MSR (R + A)- and A-histograms can be observed for a ^{252}Cf source with extending dead time values of 22.5 ns, 45 ns, 90 ns, 180 ns, and 360 ns, in Figure 1 below. The effect on both histograms is similar, the width becomes narrower and the mean of the histogram shifts to a lower neutron multiplicity value as the dead time increases. The actual dead time of the AWCC is ~ 180 ns, so we span a range.

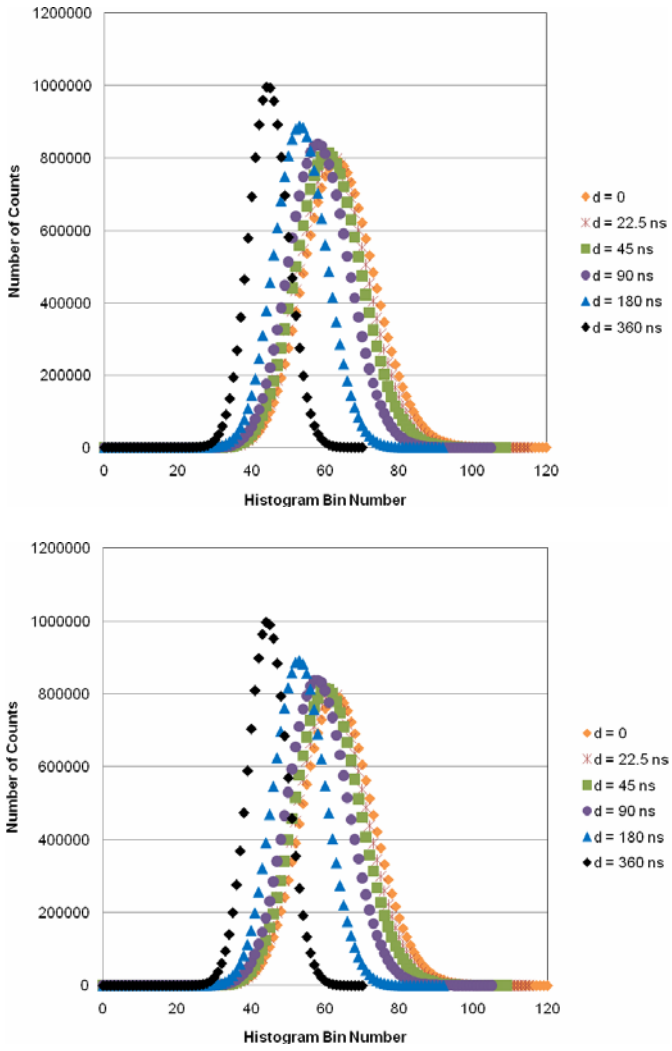

Figure 1: Case 2c ^{252}Cf. Top: (R + A)-histogram. Bottom: A-histogram.

The difference between these two histograms ((R + A) – A) can be observed in Figure 2 below, together with the effects of dead time on this quantity. The difference histogram is a measure of the number of correlated events and Figure 2 shows how the number of correlated events being detected decreases with increasing system dead time.

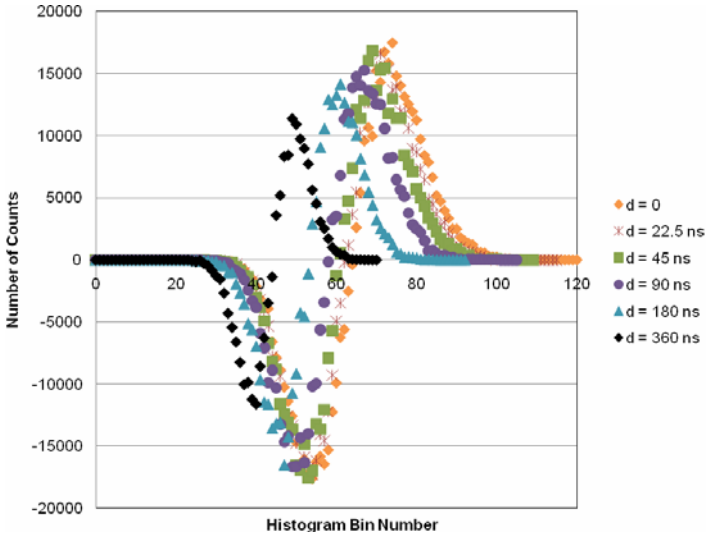

Figure 2: Case 2c ^{252}Cf. Difference ((R + A) – A) histogram

4.2. Performance comparison of selected cases

The Singles rates were corrected using the four "traditional" Singles dead time correction approaches, as well as the new Croft-Dytlewski histogram-dependent Singles rate dead time correction factor. All corrections were made using the fixed dead time value that was applied to make the pulse train i.e. parameters A and B were determined by d. The Doubles rates were also corrected using an empirical Doubles rate dead time correction factor, as well as an extension of the new histogram-dependent Singles rate dead time correction factor to derive a Doubles correction factor.

The % Recovery, given by Equation 11, was used as the performance comparison metric i.e. how well did the each of the Singles rate dead time correction factors back-correct the measured rates to the true rates. A positive result, for example, a 1 % recovery meant that the correction factors over-corrected the rates by 1 %, resulting in a corrected Singles rate 101 % of the true Singles rate value. A negative value, for example, a -1 % recovery meant that the correction factors under-corrected the rates by 1 %, resulting in a corrected Singles rate 99 % of the true Singles rate value. A zero value meant that the correction factors were able to perfectly reproduce the original true Singles rate value.

$$\%\,\text{Recovery} = \left(\frac{S_c}{S_{True}} - 1 \right) \times 100 \tag{11}$$

The % Recovery for each of the Singles rate dead time correction approaches is shown in Figure 3 as a function of the value of dead time applied. Note the relative scales: the data for the Poisson and Histogram Singles rate dead time correction factors are displayed on the same expanded scale, indicating that these two correction methods have similar performance. In the worst case, the % Recovery is -0.7 %. Similarly, the data for the Empirical and Dytlewski Trigger Singles rate dead time

correction factors are also displayed on the same scale. Here the performance is worse; the % Recovery is -10 % in the worst case, which corresponds to a 10 % under-correction.

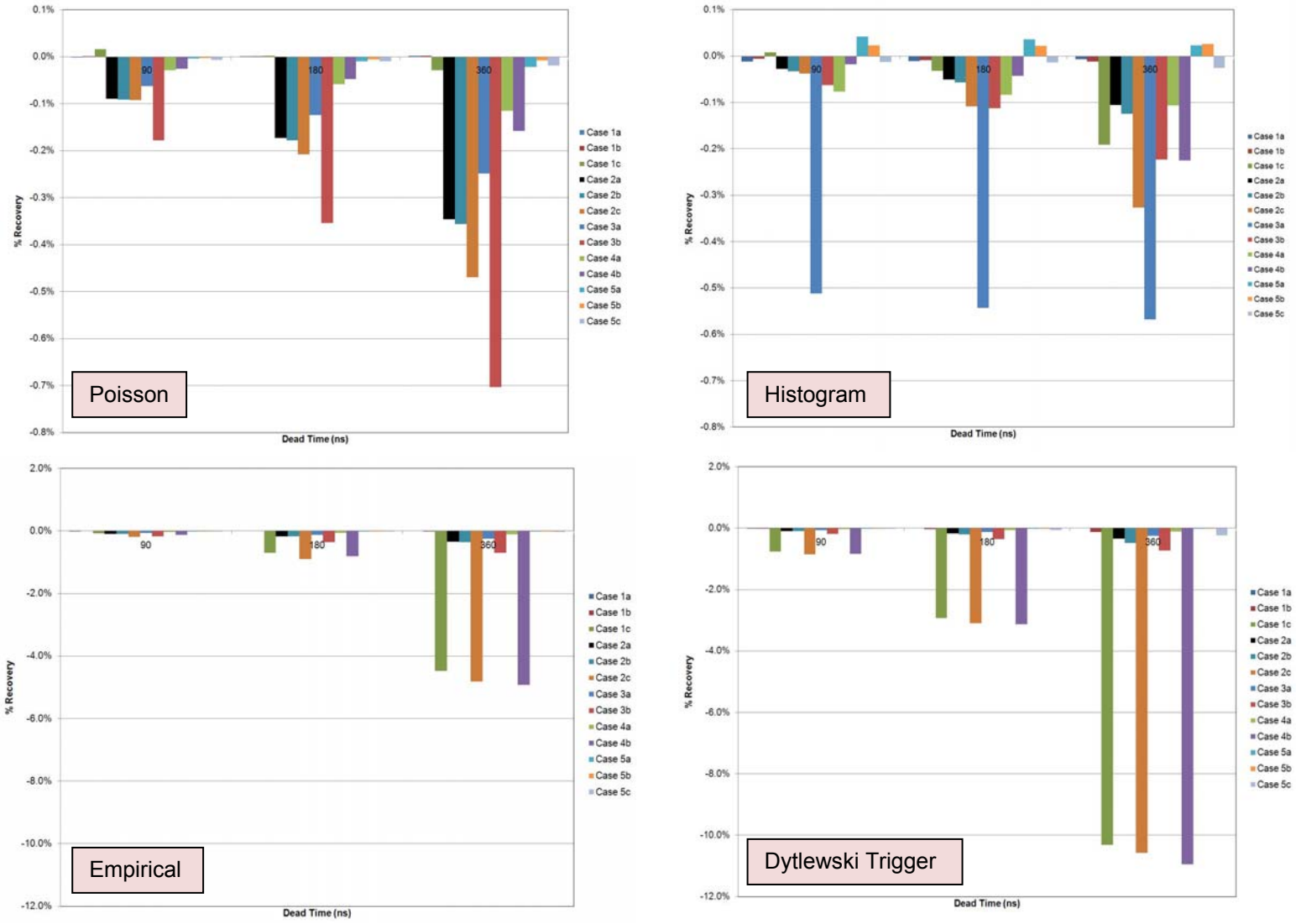

Figure 3: % Recovery performance metric for each of the four Singles rate dead time correction approaches being compared. Data is shown for all thirteen source cases at three values of input extending dead time: 90 ns, 180 ns, and 360 ns.

The % Recovery for two Doubles rate dead time correction approaches is shown in Figure 4. The top of Figure 4 shows the % Recovery for the Empirical and Histogram Doubles rate dead time correction factors. The bottom of Figure 4 shows the “Modified Histogram’ Doubles rate dead time correction, which is the same correction factor but using a higher dead time value of 220 ns in the correction factor itself (instead of 180 ns). The input extending dead time in the simulation remained at 180 ns, so the same data was re-analyzed.

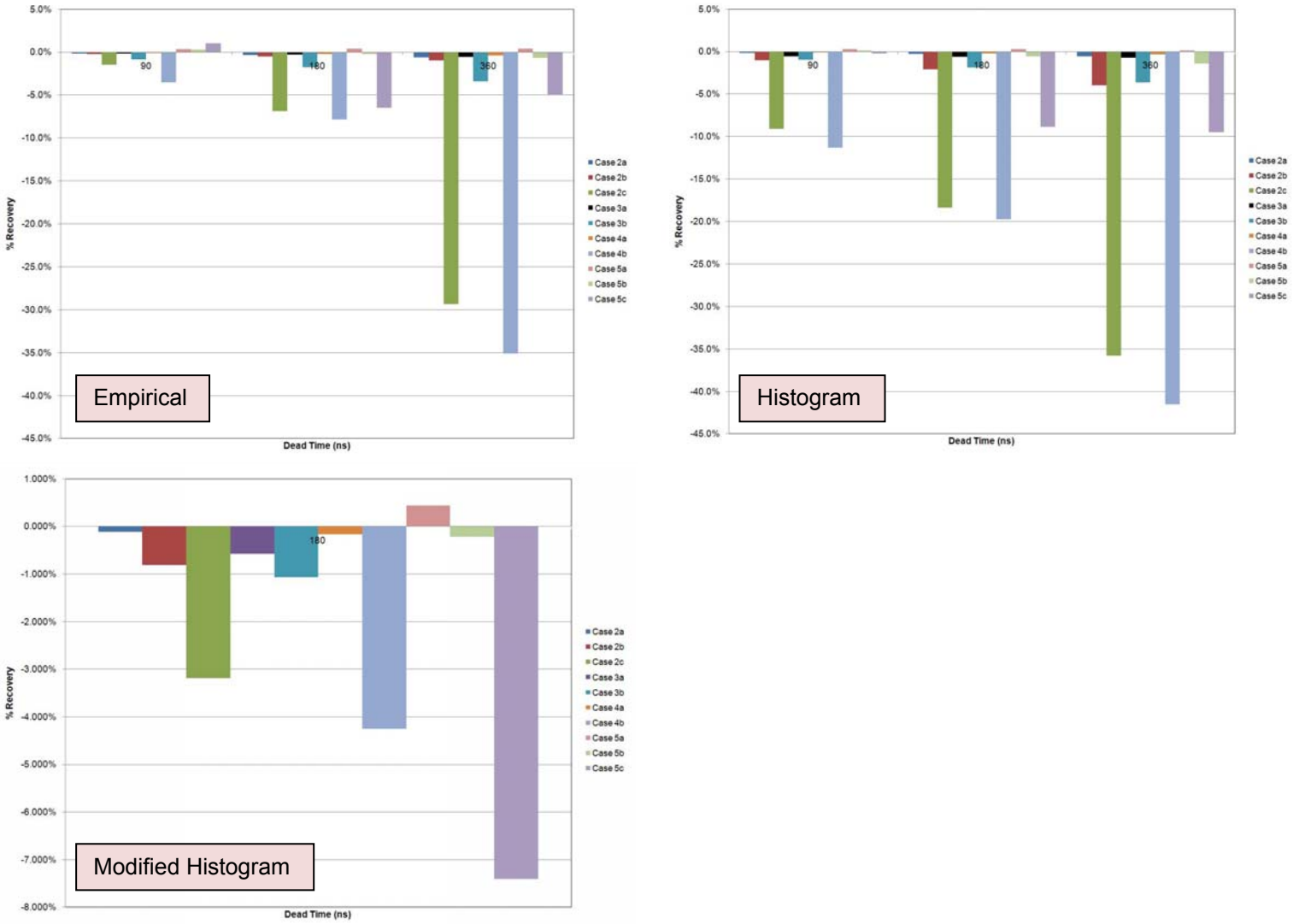

Figure 4: (Top) % Recovery performance metric for two of the Doubles rate dead time correction approaches being compared. Data is shown for ten source cases (AmLi yields only a Singles rate) at three values of input extending dead time: 90 ns, 180 ns, and 360 ns. **(Bottom)**

5. Conclusions and Future Work

We have derived alternative an alternative histogram-dependent Singles rate dead time correction factor, inspired by the Dytlewski approach. This Croft-Dytlewski dead time correction factor was tested using dead time perturbed simulated neutron pulse trains, based on the ability of the correction to recover dead time losses from the resulting histogram. All of the applied Singles rate dead time correction factors perform well for each of the AmLi source cases. This shows that the simulations of dead time effects are correct in that they truly represent an extending dead time system with a random (Poisson) neutron source. However inadequacies emerge for correlated sources. For this counter, the Poisson Singles rate dead time correction factor works well, as does the Croft-Dytlewski approach. However, the other two methods are relatively poor. Doubles corrections based on the multiplicity dead time parameter d are also poor. This emphasizes how empirical parameters are often needed to improve the correction.

Any correction method based on the correction of the MSR histograms will be item specific, since the histograms are item specific. Correction of the MSR histograms is not therefore constrained by the traditional assumptions of a true random or Poisson source. Another alternative to applying algebraic correction factors to the derived rates at the analysis stage would be to determine a method for the correction of dead time effects within the histograms themselves. Dead time corrected rates could then be derived directly from the dead time corrected histograms. This ought to allow dead time corrected correlated rates of any order to be calculated. One key research aspect of this method would be to determine the best way to "re-populate" the neutron pulse train from which the MSR histograms are derived i.e. to replace events in the pulse train that had otherwise been lost due to dead time or counting losses.

In practice, the true value of the fixed dead time is not known. It is determined by making experimental measurements and choosing a value that makes the dead time corrected count rates 'correct' at some particular counting rate. (There are several ways to determine this correct rate: twin sources, adding random counting rates to a known Doubles rate etc.) Thus it is the size of the correction that is chosen, rather than the dead time value. This approach makes a crude compensation for limitations of the chosen correction method in the range of count rates of the dead time measurement. This has worked reasonably well in many cases, but extrapolation to dissimilar counting rate regimes cannot be relied upon. The data presented here demonstrates this effect in that greatly improved results can be achieved by using the dead time corrections with a larger value for the 'dead time' than that which was applied (Figure 4). One follow-up study would be to re-create a dead time measurement (such as the twin source method) using the simulated pulse trains. The ^{252}Cf and AmLi pulse trains could be combined and the combined result re-analyzed. This would provide an indication of whether current dead time measurement methods enable the input parameters to be extracted and would expose the weaknesses in the correction methods.

The authors are preparing a more complete description of the simulation method and a "lessons learned" follow-up paper to record the potential biases in the simulation method. This paper will provide advice regarding best practice for Monte Carlo neutron pulse train simulation and post-processing in software. We shall also extend the present work to Doubles in more detail and Triples.

Acknowledgements

This work was sponsored by the U.S. Department of Energy (DOE), National Nuclear Security Administration (NNSA), Office of Nonproliferation Research and Development (NA-22).

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Front-end Electronics for Thermal Neutron Detectors

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Abstract:

Thermal neutron detectors and their associated pulse counting electronics are very critical components in safeguard measurements of fissile materials. The backbone technologies for these measurements are ^{3}He tubes and ^{235}U fission chambers, both of which require specific electronics to generate usable information. The traditional approach for the electronics has relied on the wide use of Amptek's A111 and Precision Data Technology's PDT10A preamplifier/discriminators. This approach works in many situations, but as ^{3}He and ^{235}U detectors are applied to extreme conditions (high gamma field and high count rates from spent fuel applications, long cables, high detector capacitance, low level signals from ^{235}U fission chambers, in field calibration, and the like), the limitations of the original detector design and the electronics becomes apparent.
In this paper we will discuss the factors affecting the counting performance of the detector/preamplifier system (detector response, preamplifier characteristics, artifacts due to long tails, baseline shift, etc.) and present some estimate of the magnitude of these effects. The noise aspects of the detector/preamplifier system will be discussed in light of the high capacitance and low signal level of ^{235}U chambers, long cables between the detector and electronics, and other factors.
Clearly, the traditional electronics are outdated when one considers the need to mitigate the these factors. Mitigation approaches, as well as unification of preamplifier types that can cover a variety of safeguards application, will be outlined and discussed.
Finally, methods about how to calibrate the system in the field without a radioactive source will be presented.

1. Introduction

Over the years, ^{3}He proportional counters and fission chambers have gained a reputation as the most reliable and widely used sensors for neutron measurements. The energy deposited in a ^{3}He proportional counter by a neutron reaction is above 200 keV. The counting characteristic has a well-distinguished plateau that is insensitive to threshold position (or high voltage variations) or to the shape of the neutron pulse height distribution. Fission chambers also have well-distinguished valleys between the alphas and fission fragments, but because of the lack of internal gain, they require electronics with higher sensitivity. The upcoming He-3 replacement of ^{10}B lined detectors will have overlapped neutron and gamma distributions, thus lack of real plateau of the counting characteristic. The unresolved challenges are:

- The uncertainty introduced by dead time correction increases as the count rate increases and can become the limiting factor for high accuracy multiplicity measurements
- The need to operate in harsh environments: including high humidity, wide temperature ranges, high levels of industrial noise,
- Tolerance of the electronics to the capacitance of the detector and cables.
- Spent fuel measurements: operation at high neutron and gamma rates; the gamma distribution may overlap the neutron distribution; laboratory calibration may not be accurate
- Lack of a real plateau and overlapped neutron and gamma distribution in ^{3}He replacement detectors (boron-lined)
- The threshold setting and in-field calibration procedures for the detector and electronics system typically require radiation sources and specialized operator skills. While these limitations are

acceptable in a laboratory environment, setting and re-calibrating the system in the field is much more difficult, especially in wide-scale implementation scenarios such as those for homeland security.

Many of these unresolved issues depend on signal formation in detector and signal processing in the electronics shaper/discriminator. The detector signal is well described in the text books as a convolution of two processes: the drift of primary electron inwards in the anode wire multiplication region and the much slower drift of positive ions outward the multiplication zone [1, 2]. The final signal results from another convolution between the detector signal and shaper pulse response. The amplitude and pulse height distribution of shaper signal depends strongly on the combination of the charge collection time and shaper time constant. For instance, for ^{3}He, a longer shaping time will provide a pulse height spectrum concentrated at the sum peak at 761 keV with a lower energy limit corresponding to the 191 keV of the triton.

In any shaper, the shaper response influences the signal amplitude (and thus the shape of a pulse height spectrum) as well as dead time. If the shaping time is too short, a smaller portion of the input charge pulse is integrated and therefore, due to the uncertainty in the pulse shape and duration from ^{3}He proportional counters, the pulse height distribution for a given energy will exhibit stronger variation. This corresponds to a spread in the pulse height spectrum and can cause inaccuracies in threshold determination in discriminators. To demonstrate this, Figure 1 shows pulse height spectra collected using 0.25 μs and 6 μs shaping times, which illustrates the pulse clipping. The bottom pulse height spectrum, acquired with a long shaping time, corresponds to the total integrated charge and thus exhibits the most complete features of the pulse amplitudes. The top two spectra serve to illustrate the influence of different pulse shaping (unipolar versus bipolar) on the observed pulse amplitudes. Clearly, in this case, the valley below the minimum collection energy is narrower than in a fully integrated spectrum causing possible inaccuracies in discriminator settings.

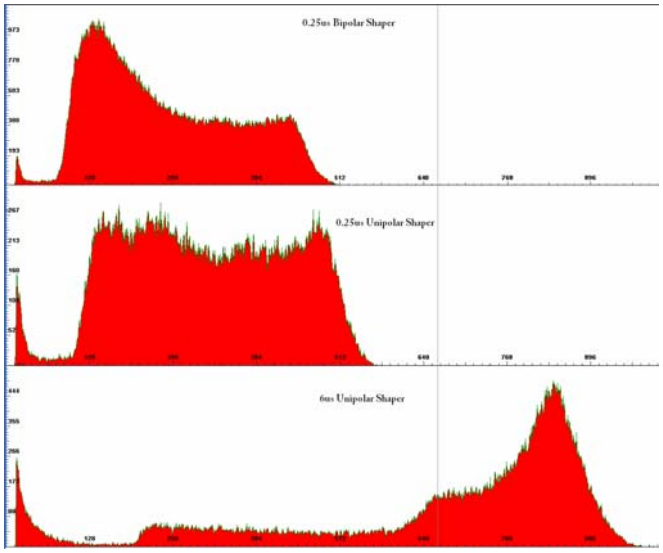

Figure 1: Pulse height spectra acquired using a ^{3}He counter with bipolar shaper with 0.25 μs shaping time (top), unipolar shaper with 0.25 μs shaping time (middle), and unipolar shaper with 6 μs shaping time (bottom).

Modeling of detector signal formation and dependence of pulse height distribution for unipolar shaper with different time constants is provided by A. Fazzi and V. Valori. [3]. While the A. Fazzi study stresses the convolution between the detector signal and different shaper responses, a more recent study lead by P.Peerani is focused on very broad and detailed characterization of various ^{3}He tubes designs and 0.5 μs unipolar shaper [4].They discuss the choice of optimal voltage to reduce space charge effects and non-linearity, the use of argon to reduce the deadtime, and the use of tailored electronics.

2. Front-end electronics

The detectors used in the thermal neutron counting and their signal parameters are summarized in table 1.

Detectors	Energy range	Charge range	Threshold setting	Pulse duration
	MeV	fC	fC	μs
^{3}He PC	0.19 – 0.760	234	14	1 - 3
^{235}U Fission Chamber	10 – 160	66	31	0.1 - 0.3
^{10}B lined PC	0 - 2.78	400	13	0.1 - 0.3

Table 1: Signal parameters of detectors used in thermal neutron counting.

The ^{3}He proportional counter (PC) detectors have a spread of pulse duration of a couple of microseconds and higher level of signals, therefore they require longer shaping times to prevent double pulsing due to pulse fluctuations. The ^{235}U fission chamber and the ^{10}B lined PC have much faster and uniform signals, thus allow shorter shaping time, but they need lower noise electronics to handle the requirement for lower threshold setting.
The front-end electronics used for neutron counting usually consists of charge (or current) sensitive preamplifier, pulse shaper, discriminator and TTL logic. This functionally is physically integrated in a small package installed directly at or nearby the detector. The pulse shaper is the most critical electronic component for neutron counting systems, determining pulse height distribution and deadtime. Almost without exception, two basic R-C shapers (unipolar and bipolar) have been used for neutron counting implementations. The timing diagram in figure 2 gives the basic properties and compares 0.5 us bipolar and unipolar shapers.

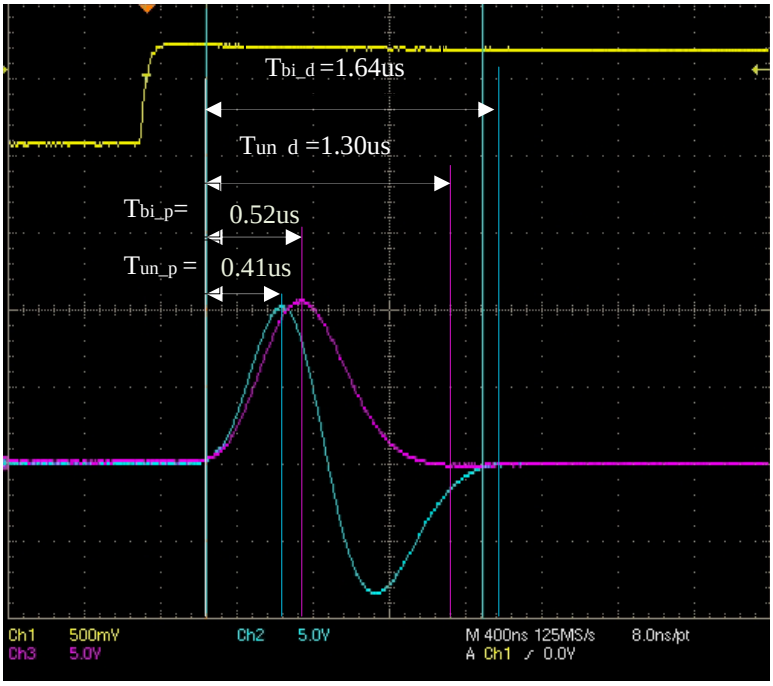

Figure 2: Shapers with unipolar and bipolar step response (400 ns/div) preamplifier pulse (yellow), bipolar shaping (magenta), unipolar shaping (purple). The time intervals for pulses' peaks and durations were measured starting from 1% of amplitude crossing until the peak maximum or the next 1% of amplitude crossing respectively.

The unipolar shaper has better filtering properties for given shaping time; the counting losses are determined only by the busy time of the discriminator. Therefore it has potentially better counting rate capabilities. The implementation is limited in the first place by the need for a gated base line restorer and difficulties in handling an intense gamma distribution and pulsed radiation field applications. In contrast the bipolar shaper has worse filtering properties for a given shaping time and more complex counting losses due to the longer time for base line recovery. Low amplitude pulses can be lost in the negative undershoot of a high amplitude pulse.
Two fast bipolar shapers have been used as a backbone of neutron counting electronics for safeguards:

- The Amptek A-111 shaper/discriminator hybrid IC, designed by Amptek Inc. [5] for operation with fast micro-channel plate detectors, was adopted by Swansen in 1984 for use in ^{3}He multiplicity counters [6]. It comprises bipolar shaping with very fast time constants which allows operation at count rates to 10^{6} cps without saturation of preamplifier section [7]. It worth mentioning that the bipolar output is not externally available which may leave the impression of unipolar shaping .
- The Precision Data Technology [8] integrated shaper-discriminator was designed to fit directly on the He3 tube. It comprises a similar bipolar shaper with a longer (about 500 ns) time constant and because of its design to fit to single detector it does not have a charge sensitive input, thus cannot tolerate more than the tube capacitance. Different packaging options of the basic shaper design are available.

The threshold setting for both PDT and Amptek A-111 shapers is based on changing the gain by a multi-turn potentiometer, which is convenient from the designer's stand point but causes setting difficulties (no reading or resetting ability). Performance evaluation of these designs can be found in [9, 10]. A recent study of the implications of front end electronics on dead time losses will be presented at the upcoming 52nd INMM meeting [11].

A fast unipolar shaper for neutron counting has been developed by A. Fazzi and colleagues [12]. Here an original implementation of gate-drain current feedback allows operation of a JFET preamplifier with a single polarity power supply. Because the drain current feedback averages the preamplifier signal, this design was implemented for coincidence counting at moderate count rates and low gamma fields [13].

Significant temperature sensitivity and high power consumption of both Amptek and PDT off the shelf designs invoked the development of a general purpose shaper discriminator described in [10]. In this paper we present some preliminary experimental results of the refined design intended to address the challenges listed above and, at the same time, maintaining versatility for use with other thermal neutron detectors.

The timing diagrams in figures 3 and 4 compare an Amptek A-111 and the LANL prototype bipolar shaper response to a standard 4 atmosphere CO_2 gas admix ^{3}He tube with a Cf-252 neutron source. Both shapers have the same duration of positive wave of shaped pulse, but the LANL shaper has less fluctuations of pulse shape and shorter undershoot because of a better shaping filter.

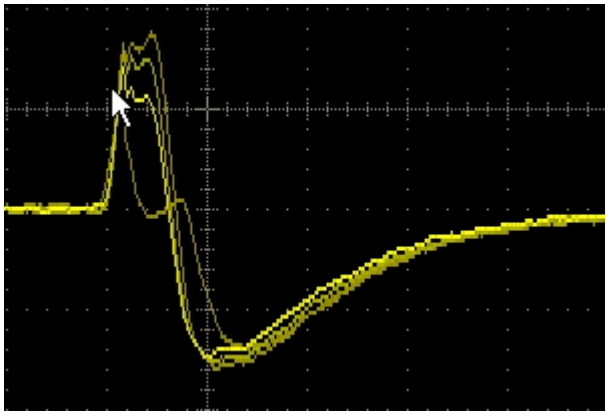

Figure 3: Amptek A-111 pulse response (signal taken from internal point). Horizontal scale 1 us/div

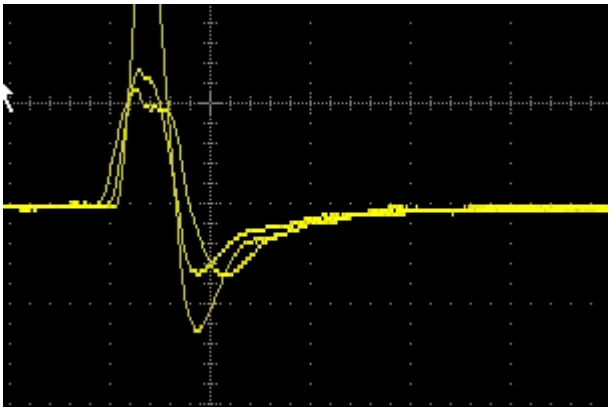

Figure 4: LANL prototype pulse response. Horizontal scale 1 us/div

The shaper prototype comprises true low noise charge sensitive input providing capability for operation with a low signal level or high capacitance detectors or long cables between the detector and shaper. The latter is a highly desirable feature for spent fuel applications where the detector can be many meters apart from the electronics. To verify the tolerance of the new design to long cables, two neutron pulse height spectra were taken with and without a 30 m long coaxial cable with 59 pf/m capacitance between the preamplifier and the detector. A reference signal (the peak above neutron distribution) from a pulser was injected for charge calibration and measurement of electronic noise. The experimental results shown in fig 5 and 6 demonstrate the tolerance to long cables or large capacitance detectors. Even given the significant broadening of reference peak, the electronic noise does not reduce the valley between the neutron and gamma distribution from the Cf-252 source. This is necessary but not a sufficient condition.

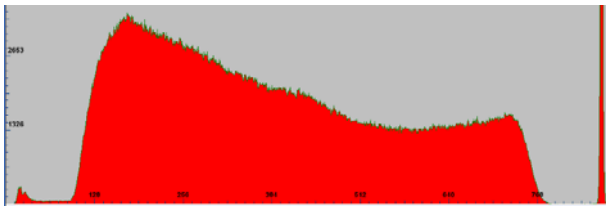

Figure 5: Pulse height spectrum without cable. The pulser peak at 300 fC position has FWHM of 1.6fC

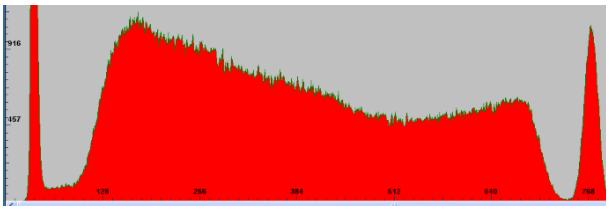

Figure 6: Pulse height spectrum with long cable. The pulser peak FWHM increased to 7.5fC

Further work for suppressing of electromagnetic interference (EMI) by using special cables or noise canceling designs as discussed in [10] will be needed to enable the desired separation between detector and electronics.

3. Spent fuel and pulsed applications

Most existing neutron measurements are at moderate count rates (up to 50 000 cps per tube) and low gamma fields. But there are some applications such as spent fuel passive and active measurements (using pulse neutron generator) where the neutron count rate will be several times higher (up to 250 000 cps per tube) and gamma field will be of the order of R/h. The space charge effect in the detector and multiple pile-up (the neutron average rate is in the ballpark of detector/electronics pulse response) could make the measurements impossible if not properly accounted. In this section we will address the gamma pile-up and space charge effects as well as optimized ^{3}He detector design.

3.1. Gamma pile-up

The gamma sensitivity of proportional counters comes from gamma interaction in the wall and the secondary electrons depositing part of their energy in detector gas [2]. The average energy deposited in the gas depends on tube size and gas stopping power [14]. The gamma distribution reduces the plateau of the counting characteristic and exceeds the neutron counting threshold at high dose rates. It is common practice to consider that gamma distribution affects the neutron distribution as a multiple pile-up of pulses of low energy. In this section we present pulse height measurements of gamma distributions and discuss the nature of gamma pile up. Figure 7 shows the whole pulse height spectrum of a 4 atmosphere ^{3}He tube with CO_2 admix taken with an unshielded Cf-252 source. Fig. 8 was taken at higher HV in order to show the valley between gamma and neutron distributions in more detail. The gamma distribution shows a well formed maximum at 15 keV corresponding to the most probable energy deposited in the gas and low intensity tail extending into the low pulse height portion of the neutron distribution.

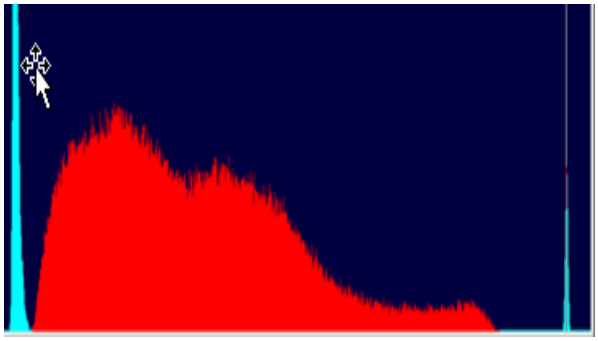

Figure 7: Pulse height spectrum with neutron and gamma distribution. LLD setting at 50 keV

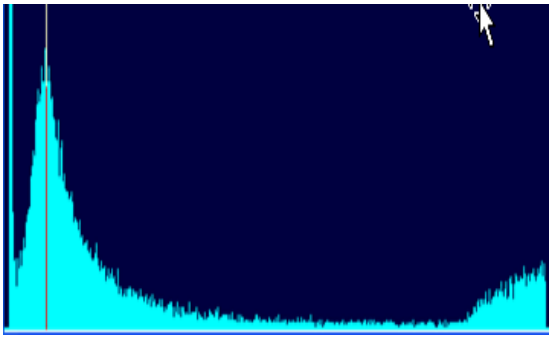

Figure 8: Pulse height spectrum of gamma distribution. (neutron distribution seen at the end of the spectrum).

This type of distribution is easy to measure at low dose rates and scale for higher dose rates. Our observations are that gammas affect the neutron distribution as single events rather than multiple pile-up. We assume that the large pulses in the tail are due to secondary electrons that travel parallel to the anode wire and can therefore have long path lengths (with corresponding low probabilities). The shape of the distribution would then be dependent on the detector geometry.

3.2. Space charge effect limitations

The counting rate dependence of proportional counter gain is well know for low gamma energy X-ray applications [14, 15]. P. Perrani and coauthors discussed and presented experimental data for space charge effect affecting the FWHM of ^{3}He tubes. We have measured the tube gain dependence of gamma pile-up by monitoring the neutron pulse height spectrum. The data reported in [10] show about 15% gain decrease at 80 R/h. The explanation of negative consequences of space charge effect is shown in conceptual plateau for different gamma dose rates (Fig. 9). Counting of gamma pulses at large values of high voltage is a well-known phenomenon. What is less well-known is that even when gamma pulses do not rise above the counting threshold, they can produce a space charge effect that reduces the gas gain in the tube. This results in a movement of the plateau characteristic to the right (higher high voltages). Therefore it can be dangerous to rely on the shape of a neutron plateau that was measured in the absence of gamma dose in order to determine the actual measurement efficiency when a significant gamma dose rate is present.

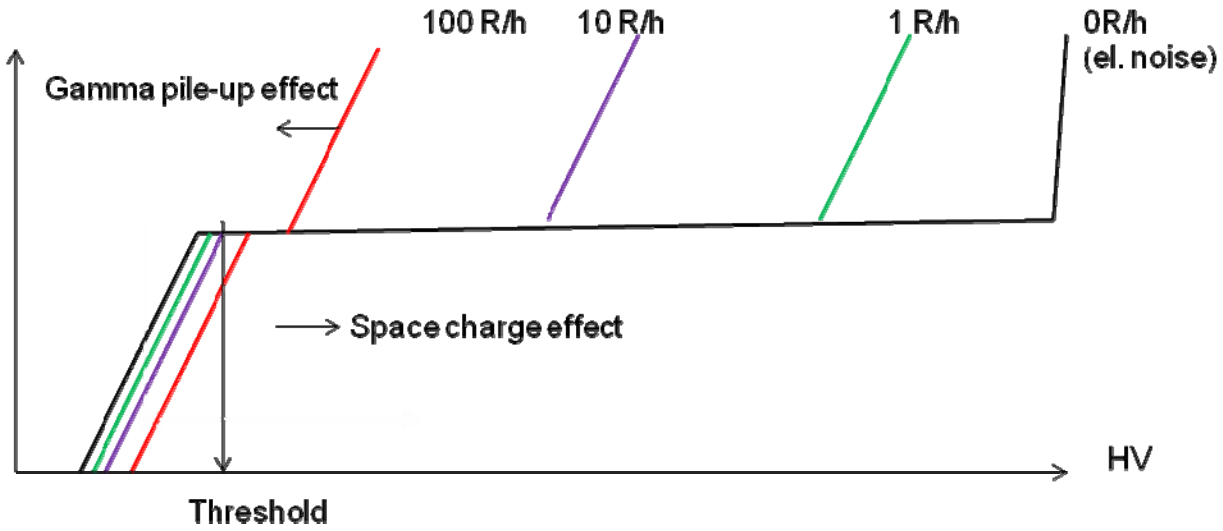

Figure 9: The charge effect shifts the knee of plateau characteristic toward higher voltage , therefore it can set the threshold below the knee , thus change the calibration.

While the space charge effect from gamma is known, there are no data for space charge effects at high neutron rates. To prevent this undesirable effect, one has to operate the tubes at lower gain or use tube designs resilient to space charge effects. A good practice would be to characterize the tube space charge boundaries and monitor the average tube current during all measurements with potentially hot samples.

4. Optimization of ^{3}He tube design for high count rate and pulsed applications

The ^{3}He proportional counter is a very mature technology with geometry design dominated by 0.002" anode wire and 1" tube diameter. Most of the optimization efforts are based on selection of gas pressure and gas admix as a tradeoff between gamma sensitivity and dead time [16] or optimization of HV settings [4]. Here we explore another optimization option: the geometry of the electrical field. The space charge effect suggests operation of the tube at voltages below the commonly used values. Experimental data presented by P. Perrani for tubes with 0.001" and 0.002" diameters and identical gas fill show higher operating voltage and better dead time for the tube with higher anode wire diameter. We took more extreme approach and increased the tube wire diameter to 0.004". The operating voltage for the tube to have the same gain increased by a factor of two and the spread of charge collection times decreased by about of factor of two. The timing diagram for differentiated charge pulses are shown in fig. 10 and fig. 11 .The lower rise time spread of for a tube with a thicker wire allows shorter shaping times to be used. Additional improvement can be achieved by tailoring the shaping to the detector pulse response and including compensation of the longer time ionic component.

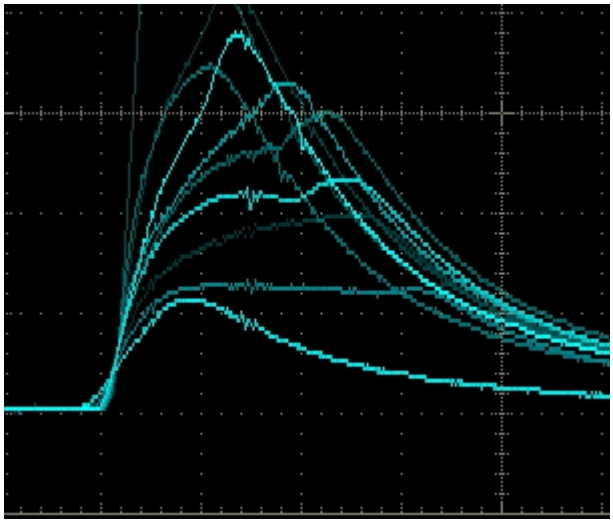

Figure 10: Pulses timing diagram for tube with 0.002" anode wire. (horizontal scale 400 ns/div)

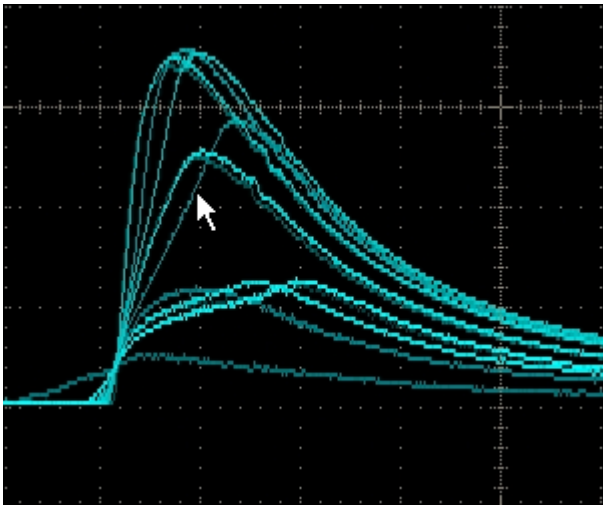

Figure 11: Pulses timing diagram for tube with 0.004" anode wire. (horizontal scale 400 ns/div)

Because the counting conditions in pulsed applications change dramatically, the space charge effect and the dead time of the detector and electronics change the neutrons count rate time profile especially when the neutron signal has maximal intensity. The tube and electronics has to operate during the flash or recover very quickly after the initial flash or burst of neutrons. Operating the detector at lower gain is critical for avoiding the screening effect of space charge that can last many microseconds [15]. Operating the tubes at lower gain or the use of tubes with more uniform distribution of electrical field (thicker anode designs) would avoid nonlinear effects and enable operation right after the flash. Unipolar shapers with their long time constants for base line recovery are not the preferred choice for front-end electronics with fast recovery. The LANL developed prototype is intended to recover within a few microseconds after the flash without the need for gating.

5. Acknowledgements

This work by large was accomplished under internal LANL Program Development project for advanced detectors and electronics.
We would like to thank Eric Smith for useful discussions on space charge effects and deadtime and Amptek Inc. for information on the operation of the A-111 amplifier.

6. Conclusion

A new preamplifier/shaper design has been presented that is suitable for ^{3}He tubes, boron lined detectors and fission chambers, all operating at high count rates.
Space charge effects in proportional counters were discussed in light of changing tube gain and introducing possible errors in efficiency calibration.
The pulse height distribution of gamma pile-up was discussed. The observed gamma pile-up was explained as single events rather than multiple low amplitude events. This has implications for the design of tubes, the design of processing electronics and also for simulations of gamma effects.
Improvement of ^{3}He dead time and space charge tolerance was proposed by the use of a more uniform distribution of electric field by using a thicker diameter anode wire. In order to obtain the full benefits of this change, it is necessary to use tailor-made electronics to compensate for the slow tail of the ionic component of the signal.

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On the Feynman-alpha formula for fast neutrons

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Abstract

In this contribution, a stochastic theory for a branching process in a neutron population with two energy levels is investigated. In particular, a variance to mean or Feynman-alpha formula is derived in this generalized scenario using the Kolmogorov forward or master equation theory for the probabilities in a system with a compound Poisson source.

1 Introduction

There exist several relatively new applications where the energy distribution of the neutrons plays a significant role. One particular case is a method used in nuclear safeguards, namely the stochastic generalization of the so-called differential die-away analysis (DDAA) [1, 2, 3, 4]. Traditionally, the DDAA method was used as a deterministic method of detecting fissile material embedded in moderating surroundings using a pulsed source. The newly explored method, called differential die-away self-interrogation (DDSI) utilizes the inherent spontaneous neutron emission of the sample [5]. In the DDSI method the temporal decay of the correlations as a function of the time delay between two detections of fast neutrons is used. This corresponds to a Rossi-alpha measurement with two energy groups. Furthermore, in recent pulsed experiments measuring the reactivity in fast cores of accelerator driven sub-critical systems it is found that two exponentials appear, indicating that the temporal behavior of the fast and thermal neutrons is separated in fast reflected cores. This leads to the fact

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that a two group versions of the Feynman and Rossi-alpha formulas are needed [6]-[7].

In this contribution, a stochastic theory for a branching process in a neutron population with two energy levels is investigated based on the previous results in Refs [6]- [7]. In particular a variance to mean or Feynman-alpha formula is derived in this generalized scenario using the master equation or Kolmogorov forward approach. The model includes absorbtions, down scattering from fast to thermal neutrons, thermal fissions, detections and an external source of fast neutrons. Higher moments will also be discussed as well as specific applications to areas within safeguards research as well as specific applications to areas within safeguards research.

2 The variance to mean via the forward Kolmogorov approach

In this section we will describe the two particle type system by using the Kolmogorov forward approach. We will include a compound Poisson source of fast neutrons described by the source strength S_1 which releases n particles with probability $p_q(n)$ at an emission event (i.e. spontaneous fission). The source is assumed to be switched on at time $t = t_0$, although dependence on t_0 will not be denoted. The effects of detecting particles will also be included, denoted by the intensity λ_d. We will start by giving the differential equation for the probability $P(N_1, N_2, Z_1, t)$ for N_1 fast, N_2 thermal neutrons at time t and Z_1 detected fast particles in the interval $(0, t)$. We have summed all mutually exclusive events during an infinitesimally

small time interval dt and find for probabilities,

$$\begin{aligned}\frac{\partial P(N_1,N_2,Z_1,t)}{\partial t} &= -(\lambda_1 N_1+\lambda_2 N_2+S_1)P(N_1,N_2,Z_1,t)\\ &+ \lambda_{1a}(N_1+1)P(N_1+1,N_2,Z_1,t)\\ &+ \lambda_{2a}(N_2+1)P(N_1,N_2+1,Z_1,t)\\ &+ \lambda_R(N_1+1)P(N_1+1,N_2-1,Z_1,t)\\ &+ \lambda_{2f}(N_2+1)\sum_k^{N_1} f(k)P(N_1-k,N_2+1,Z_1,t)\\ &+ \lambda_d(N_1+1)P(N_1+1,N_2,Z_1-1,t)\\ &+ S_1\sum_n^{N_1} p_q(n)P(N_1-n,N_2,Z_1,t). \end{aligned} \quad (1)$$

Here, λ_1 and λ_2 are the decay constants (total reaction intensities) for fast and thermal neutrons whereas λ_{1a}, λ_{2a} are the absorbtion intensities of fast and thermal particles, respectively. The removal of fast particles into the thermal group is described by λ_R while fission resulting from the thermal particles happens with the intensity of λ_{2f}. The intensities are related through

$$\lambda_1 = \lambda_{1a}+\lambda_R+\lambda_d, \quad (2)$$

and

$$\lambda_2 = \lambda_{2a}+\lambda_{2f}. \quad (3)$$

We will now solve this differential equation by using the generating function of the form

$$G(X,Y,Z,t) = \sum_{N_1}\sum_{N_2}\sum_{Z_1} X^{N_1}Y^{N_2}Z^{Z_1}P(N_1,N_2,Z_1,t), \quad (4)$$

and describe the process in the time evolution of the generating function as,

$$\begin{aligned}\frac{\partial G}{\partial t} &= (\lambda_{1a}+\lambda_R Y+\lambda_d Z-\lambda_1 X)\frac{\partial G}{\partial X}+(\lambda_{2a}+\lambda_{2f}\nu(X)-\lambda_2 Y)\frac{\partial G}{\partial Y}\\ &+ S_1(r(X)-1)G, \end{aligned} \quad (5)$$

where

$$\nu(X) = \sum_k f_k X^k, \qquad \text{and} \tag{6}$$

$$r(X) = \sum_n p_q(n) X^n. \tag{7}$$

Here, f_k is the probability of having exactly k neutrons produced in a fission event. Differentiation of equation (5) with respect to (X, Y, Z) and then letting $(X = Y = Z = 1)$ yields differential equations for the expectations as,

$$\frac{\partial}{\partial t}\langle N_1\rangle = -\lambda_1\langle N_1\rangle + \lambda_{2f}\nu_1\langle N_2\rangle + S_1 r_1, \tag{8}$$

$$\frac{\partial}{\partial t}\langle N_2\rangle = -\lambda_2\langle N_2\rangle + \lambda_R\langle N_1\rangle, \tag{9}$$

$$\frac{\partial}{\partial t}\langle Z_1\rangle = \lambda_d\langle N_1\rangle. \tag{10}$$

Here we have used the definition of the derivatives on the equations (6) and (7) as $\nu_1 = dq/dX|_{X=1}$ and $r_1 = dh/dX|_{X=1}$. We note that due to the source term with intensity S_1 the dynamical system consisting of equations (8) - (9) will reach a steady state ($\frac{\partial}{\partial t} \to 0$) and we find the stationary solution,

$$\langle N_1\rangle = \bar{N}_1 = \frac{\lambda_2 S_1 r_1}{\lambda_1\lambda_2 - \nu_1\lambda_R\lambda_{2f}} = \frac{\lambda_2 S_1 r_1}{\omega_1\omega_2}, \tag{11}$$

$$\langle N_2\rangle = \bar{N}_2 = \frac{\lambda_R S_1 r_1}{\omega_1\omega_2}, \tag{12}$$

$$\langle Z_1\rangle = \epsilon\lambda_{2f}\bar{N}_1 t, \tag{13}$$

where $\epsilon = \lambda_d/\lambda_{2f}$ and we have used the additional definitions ω_1 and ω_2,

$$-\omega_1 = -\frac{1}{2}(\lambda_1 + \lambda_2) + \frac{1}{2}\sqrt{(\lambda_1 - \lambda_2)^2 + 4\lambda_1\lambda_2\nu_{eff}}, \tag{14}$$

$$-\omega_2 = -\frac{1}{2}(\lambda_1 + \lambda_2) - \frac{1}{2}\sqrt{(\lambda_1 - \lambda_2)^2 + 4\lambda_1\lambda_2\nu_{eff}}, \tag{15}$$

$$\nu_{eff} = \nu_1\frac{\lambda_R\lambda_{2f}}{\lambda_1\lambda_2}. \tag{16}$$

The expectation of the detections is found by integrating equation (10) and we note that the number of detections increases linearly with time. In order

to find the variance of the detector counts we need to determine the second factorial moment by yet another differentiation with respect to (X, Y, Z) and then letting $(X = Y = Z = 1)$. The variance of the detector counts can be determined through the relation $\sigma_Z^2 = \langle Z_1 \rangle + \mu_{ZZ}$ where the modified variance μ_{ZZ} is defined as $\mu_{ZZ} = \langle Z(Z-1) \rangle - \langle Z \rangle^2 = \sigma_{ZZ}^2 - \langle Z \rangle$ while in general we have $\mu_{XY} = \langle XY \rangle - \langle X \rangle \langle Y \rangle$. The differentiation procedure gives a system of six dynamical equations of the modified second moments as

$$\frac{\partial}{\partial t}\mu_{XX} = -2\lambda_1 \mu_{XX} + 2\nu_1 \lambda_{2f} \mu_{XY} + \nu_2 \lambda_{2f} \bar{N}_2 + S_1 r_2, \tag{17}$$

$$\frac{\partial}{\partial t}\mu_{XY} = -(\lambda_1 + \lambda_2)\mu_{XY} + \lambda_R \mu_{XX} + \nu_1 \lambda_{2f} \mu_{YY}, \tag{18}$$

$$\frac{\partial}{\partial t}\mu_{YY} = -2\lambda_2 \mu_{YY} + 2\lambda_R \mu_{XY}, \tag{19}$$

$$\frac{\partial}{\partial t}\mu_{ZX} = -\lambda_1 \mu_{ZX} + \nu_1 \lambda_{2f} \mu_{ZY} + \lambda_d \mu_{XX}, \tag{20}$$

$$\frac{\partial}{\partial t}\mu_{ZY} = -\lambda_2 \mu_{ZY} + \lambda_R \mu_{ZX} + \lambda_d \mu_{XY}, \tag{21}$$

$$\frac{\partial}{\partial t}\mu_{ZZ} = 2\epsilon \lambda_{2f} \mu_{XZ}, \tag{22}$$

where we have used the additional notations $\nu_2 = d^2q/dX^2|_{X=1}$ and $r_2 = d^2h/dX^2|_{X=1}$. Although, the system of equations (17) - (22) is rather complicated and an analytical solution would be hard to find, we note that in stationary state the system breaks down into two systems independent of each other where the moments $\mu_{XX} = \bar{\mu}_{XX}$, $\mu_{XY} = \bar{\mu}_{XY}$ and $\mu_{YY} = \bar{\mu}_{YY}$ are constants. However, the equations describing detected particles need to be solved by e.g. Laplace transforms of (20) and (21) whereas it is possible to find the sought moment μ_{ZZ} by integration by using equation (22). We find the constant 2nd modified moments as,

$$\bar{\mu}_{XX} = \frac{(\lambda_2^2 + \omega_1\omega_2)(\nu_2 \lambda_{2f} \bar{N}_2 + S_1 r_2)}{2(\lambda_1 + \lambda_2)\omega_1 \omega_2}, \tag{23}$$

$$\bar{\mu}_{XY} = \frac{\lambda_2 \lambda_R (\nu_2 \lambda_{2f} \bar{N}_2 + S_1 r_2)}{2(\lambda_1 + \lambda_2)\omega_1 \omega_2}, \tag{24}$$

$$\bar{\mu}_{YY} = \frac{\lambda_R^2 (\nu_2 \lambda_{2f} \bar{N}_2 + S_1 r_2)}{2(\lambda_1 + \lambda_2)\omega_1 \omega_2}. \tag{25}$$

The objective now is to solve (20) and (21) by Laplace transform methods and we find the transformed identity as,

$$\tilde{\mu}_{XZ} = \frac{\nu_1 \lambda_d \lambda_{2f} \bar{\mu}_{XY}}{sH(s)} + \frac{(s+\lambda_2)\lambda_d \bar{\mu}_{XX}}{sH(s)} \tag{26}$$

with

$$H(s) = s^2 + (\omega_2 + \omega_1)s + \omega_1\omega_2. \tag{27}$$

Note that we have assumed that the initial values of the moments μ_{XZ} and μ_{YZ} were equal to zero at $t = 0$ (at the start of the measurement), hence the roots of $H(s)$ determine the temporal behavior of the Feynman-alpha formula. Moreover, the solution has many similarities to that found in Ref. [8]. The variance $\sigma_{ZZ} = \langle Z \rangle + \mu_{ZZ}$ is now found by integration of (22) and after some algebra the Feynman-alpha formula can now be written in the form

$$\frac{\sigma_{ZZ}(T)}{Z_1} = 1 + Y_1(1 - \frac{1 - e^{-\omega_1 T}}{\omega_1 T}) + Y_2(1 - \frac{1 - e^{-\omega_2 T}}{\omega_2 T}). \tag{28}$$

Here, the complete expressions for Y_1 and Y_2 are quite lengthy. However, it turns out that the sum $Y_0 = Y_1 + Y_2$ takes a rather simple form that also determines the value of the Feynman-alpha for large measurement times $T \to \infty$ as,

$$Y_0 = Y_1 + Y_2 = q_2 \frac{\lambda_d \lambda_2 \lambda_R \lambda_{2f}}{\omega_1^2 \omega_2^2}. \tag{29}$$

We will now turn our attention to some quantitative examples of the Feynman-alpha formula in the form of Equation (28).

3 Results and discussion

The Feynman-alpha formula for a two particle type system found by using the Kolmogorov forward approach including a Poisson source and effects of detecting particles is shown in Figure 1 (A and B). We have used the parameters $\nu_1 = 3.0$, $\nu_2 = 5.0$, $S_1 = 1.0$, $r_1 = 1.0$, $r_2 = 0.0$, $\lambda_{2f} = 3/5$ and $\lambda_d = 0.1$. In Figure 1A $\lambda_R = 2/3$ whereas in Figure 1B $\lambda_1 = 1.0$ and $\lambda_2 = 2.0$. As expected, the curves grow according to Equation (28) exponentially

to a maximum value determined by the constant Y_0. However, this value can signifcantly vary depending on the intensities (λ_1, λ_2, etc) involved the process. The increase of the curves is determined by two exponentials. Furthermore, unlike the case of the DDSI method, the presence of the two exponentials is not visible to the bare eye. In Figure 1A, it is seen that the ratio of the decay intensities for fast and thermal particles has a nontrivial effect on the maximum value by changing the ratio in the range ($0.25-4.0$). In Figure 1B, the effect of the thermalization process described by the intensity λ_R on the results is illustrated. Increasing thermalization increases the asymptotic value of the Feynman-alpha.

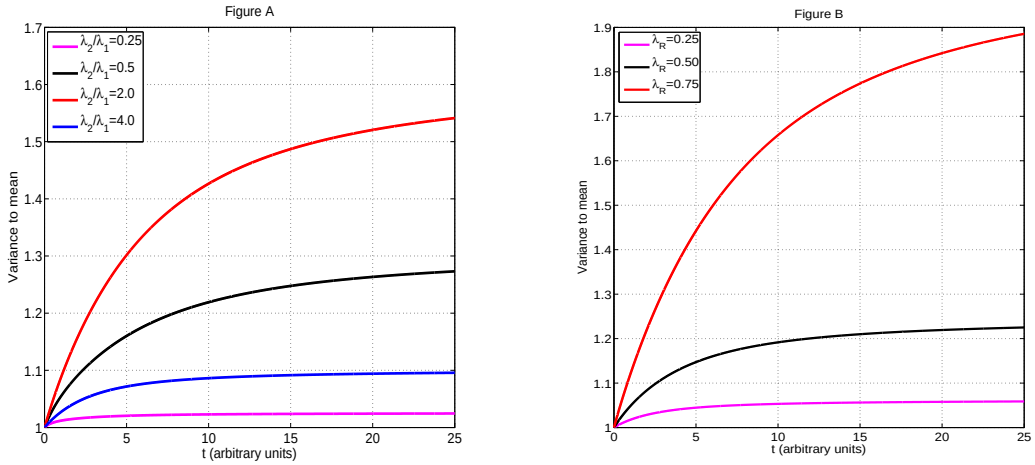

Figure 1: (A and B) The Feynman-alpha expression is shown for the parameters $\nu_1 = 3.0$, $\nu_2 = 5.0$, $S_1 = 1.0$, $r_1 = 1.0$, $r_2 = 0.0$, $\lambda_{2f} = 3/5$ and $\lambda_d = 0.1$. In Figure A, $\lambda_R = 2/3$ whereas in Figure B $\lambda_1 = 1.0$ and $\lambda_2 = 2.0$.

4 Conclusions

We have developed a forward Kolmogorov approach for the two group theory of the Feynman-alpha method, including a compound Poisson source and the detection process. The results agree with those calculated by the backward approach as reported in [6]. It is seen that, unlike in the DDSI

method (i.e. the two-group version of the Rossi-alpha method), the presence of two exponents in the solution is not clearly visible. This means that detection of the presence of fissile material may not be as obvious as with the Rossi-alpha method. On the other hand, the determination of the exponents ω_1 and ω_2 by curve fitting could be more accurate in certain cases than with the DDSI method. However, the diagnostic value of the exponents in terms of determination of the sample parameters is not clear yet, and it requires further investigations, which will be reported in further work.

5 Acknowledgements

This work was supported by the Swedish Radiation Safety Authority (SSM).

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19 Non-proliferation technical aspects

Applicability of the Directed Graph Methodology for Acquisition Path Analysis

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Abstract:

Possible methods to construct, visualize and analyse the "map" of the State's nuclear infrastructure based on different directed graph approaches are proposed. The transportation and the flow network models are described in detail. The use of the possible evaluation methodologies and the use of available software tools to construct and maintain the nuclear "map" using pre-defined standard building blocks (nuclear facilities) are introduced and discussed.

Keywords: acquisition path analysis; directed graph; shortest path algorithm; State level approach; transport model; material flow network model

1. Introduction

The State-level approach to IAEA safeguards requires the detailed knowledge and objective evaluation of a State's nuclear infrastructure, including human and economical resources. One possible outcome of such an evaluation is the possible nuclear material diversion roots or paths required for the acquisition of the first nuclear weapon or nuclear explosive device. The technical basis of the evaluation is the physical model of the nuclear fuel cycle, which was developed by the IAEA [1].

Recently (in 2010) the IAEA requested Member State support in the development of acquisition path analysis methodology and applicable software tools. In response to the request, the paper discusses the possible use of two different analytical approaches based on graph models: the shortest path algorithm applied to a transportation type model and the material flow network. Both models are discussed in three different levels.

2. Modelling approaches

Both models are based on a network. A network might be defined as any system of interconnected linear features. A network can be composed of nodes and links between the nodes (edges). In mathematical sense these types of networks are called "graphs" that are mathematical structures used to model pair wise relations between objects from a certain collection. A graph may be undirected, meaning that there is no distinction between the two vertices associated with each edge, or its edges may be directed from one vertex to another.

The processes in a nuclear fuel cycle are either irreversible ones (e.g. irradiation of fuel elements in a reactor) or directed in practice (conversion of materials). Therefore, the two modelling approaches proposed in this paper are directed graph models.

2.1. Acquisition Path Analysis and the Shortest Path Problem (Transport modelling)

In graph theory, the shortest path problem is the problem of finding the shortest path between two selected nodes – the source and the destination – of the graph in such a way that the length of the path – the sum of the weights assigned to the edges belonging to the path – is the shortest of all existing paths [2]. The length of this shortest path is called the distance of the two nodes.

The problem has several forms which are various generalizations of the simple case. These are as follows (including the basic case):

- "single-pair problem": in this basic case as described above, the task is to find the shortest path connecting a selected pair of nodes, the source and the destination;
- "single-source problem": here a single source is selected and the shortest paths from a the source to all other nodes (as destinations) are to be found;
- "single-destination problem": the reverse of the "single-source problem", to find the shortest paths from all nodes as sources to a selected destination (this is identical to a "single-source problem" where the source is replaced with the destination and the edges are reversed);
- "all-pairs problem": in this "full" version of the problem the shortest path between all (connected) pairs of nodes have to be found (obviously, the solutions of the three previous cases are a subset of the solutions of the "full" analysis).

There are various well documented algorithms for the solution of all four types of problems[1]. Without going into the computational details, it is worth to note, that the required computation time is a polynomial function of the number of nodes and edges, i.e. the algorithms are efficient and large complex graphs with thousands of nodes and edges can be analysed without computational difficulty.

A simple – and probably the best known – application of the "single-pair" shortest path problem can be found in the various existing route planning software programs. Here the task is to find the shortest way on a map from one town (or in general from a geographical location) to another. Here the graph is the map with the towns (locations) as nodes and the roads connecting the towns being the edges. If the weights of the edges are the physical lengths of the connecting roads, the resulting distance is the physical distance of the locations along the real road. However, the weights might be assigned different values: time needed to cover the segment of the road, the cost of fuel consumed plus road toll, etc., or a suitable combination of these. Consequently the resulting shortest path solution might in reality be the quickest or the cheapest way to get from one location to the other or an optimal route based on a combination of the features taken into consideration in the computation of the weights. Furthermore, in a route planning application one can select what kinds of roads (motor way, country road, bridge, ferry) to use or not to use (in general: to set an order of preference of these) and what towns to visit or to avoid. As clearly seen on the example of the route planning applications, the shortest path approach offers a great flexibility of analysing complex graphs of thousands of nodes and edges (towns and roads on the map) from different points of view, depending on how the weights assigned to the edges are defined.

Using the analogy of route planning, if the nuclear fuel cycle is described as a map (graph, flow-chart, process-diagram), the acquisition path analysis task can be formulated as a shortest path problem, where the nodes are the different stages of the fuel cycle (facility types, material types), the edges describe possible transitions between them, and the destination is the nuclear weapon (actually, there are three separate destinations which give rise to three, independent problems: one for the ^{235}U bomb, one for the Pu bomb and the third one for the ^{233}U bomb.

Running a "single-pair" shortest path algorithm between any selected node and the destination will give the optimal sequence of steps from that node (stage of the fuel cycle) to reach the destination (nuclear weapon) and establish the lower bound on the length, i.e. the distance. Similarly, running a "single-destination" algorithm for the same graph will provide the results for all nodes as source points in one go. Of course the meaning of the distance will depend on how the weights of the edges are assigned.

[1] The simplest algorithm for the solution of the "single-pair" and "single source/destination" problem is Dijkstra's algorithm. Any single-pair algorithm can be used for the solution of the generalized cases by simply running the "single-pair" algorithm for all selected pairs. However, more efficient algorithms were developed for the solution of the "single source/destination problem" (e.g. the Bellman-Ford algorithm) and of the "all-pairs problem" (e.g. the Floyd-Warshall or the Johnson algorithm).

2.2. Material flow network modelling

Another way of representing the nuclear profile of a State is to model the theoretical performance capacity of its nuclear infrastructure by the possible ways of producing nuclear materials suitable for manufacturing a weapon from nuclear and other materials at its disposal. This approach can be formulated via a flow network model.

Materials in different chemical and physical form can flow through pipes representing actual processes in the nuclear fuel cycle. Any material flowing through the network can only change its state and quantity by passing through one of the pipes. A given pipe corresponds to an actual process in a given facility type having specific input and output material type. The length of a pipe represents the time needed to process a unit mass of the input material, while the cross section of the pipe scales with the capacity of the process. Import and export of materials can also be taken into account by pipes. A state parameter having value between 0 > – 1 is also assigned to each pipe giving the probability (or possibility) of the existence of the process. A pipe in state 1 is a declared process, while in state 0.1 is an undeclared process which exists with a probability of 0.1.

Each node of the network represents the amount of a material type in a given chemical and physical form present in the State. A node can be composed of sub-nodes representing sub-amounts of material that is actually in the same physical place in a State if needed, but this is irrelevant from the state level evaluation point of view.

As an illustration, two possible pipes are shown in Fig 1., while a possible subset of a flow network is illustrated in Fig. 2.

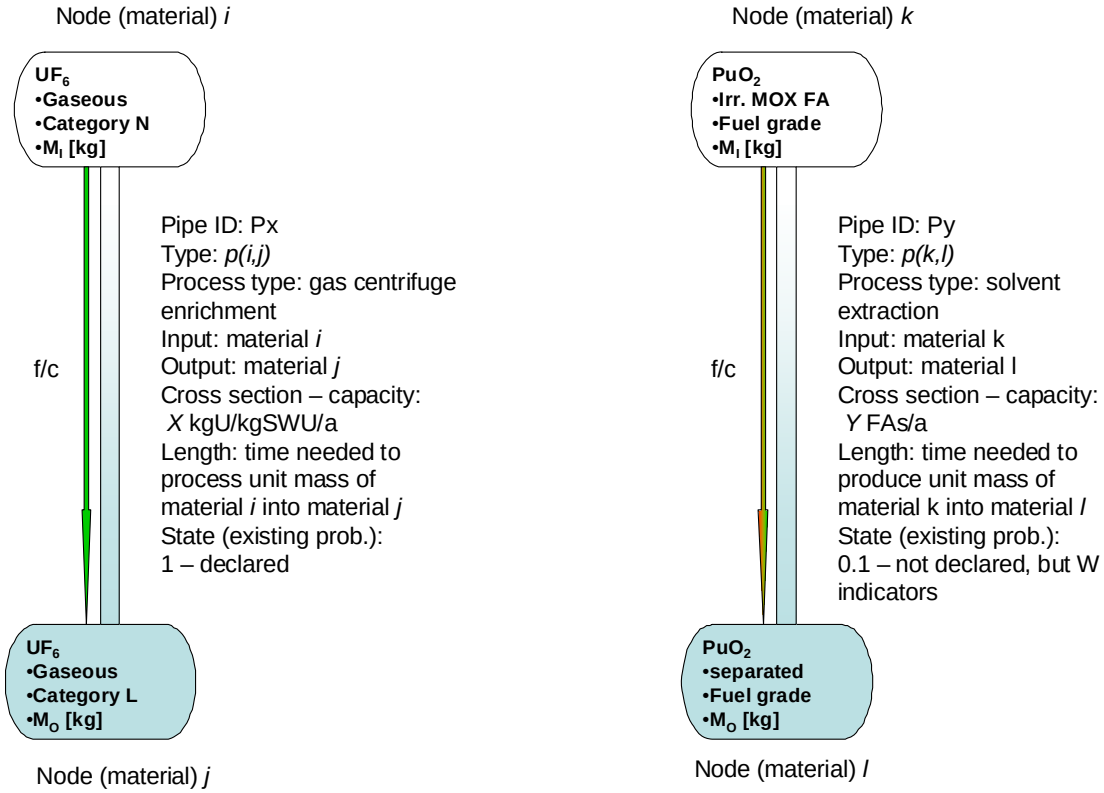

Figure 1. illustrative elements of a flow network of a State's nuclear capacity

In Fig. 1 the left pipe represent a gas centrifuge enrichment process with natural UF_6 gas as input and low enriched UF_6 as output, with technical details as declared and verified. The right pipe is an undeclared reprocessing process based on solvent extraction with irradiated MOX fuel as input and separated PuO_2 as output with existence probability of 0.1 that is based on the presence of weak indicators of the process.

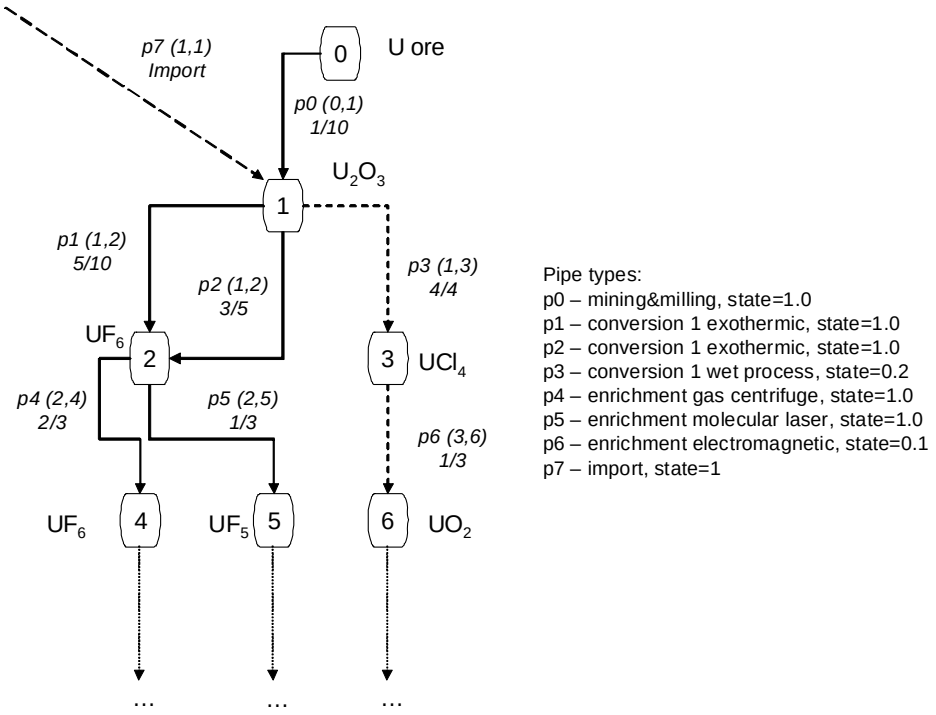

Figure 2. Possible subset of a material flow network

In Fig. 2, seven nodes and seven processes are indicated representing a frontend part of the material flow network of the fuel cycle.

The material flow network model can be mathematically described as a finite directed graph G(V,E) with every edge (pipe) $p(i,j) \in E$ having positive capacity (non-zero cross section), finite length and positive state parameter with maximum value of 1. An edge $p(i,j)$ usually connects two different nodes i and $j \in V$ (a pipe processing material i into j with a given flow and capacity). An edge however can also connect i to i . In this case the edge represents an outside source or sink of material i (import or export for instance). There are target nodes $t \in V$ in the network as elements of a set of nodes $T \in V$, representing weapons grade materials.

A path $P(s,t)$ in the network connecting a non target node *s* to target node *t* exists if there are successive edges starting from *s* and leading to *t* with positive flow. Similarly to the state value of the edges, a path can have a probability < 1, if it contains edges with a probability < 1.

There are several possibilities to formulate a problem to be solved on this type of flow network. The most obvious one would be to find all the possible paths in the network that can lead to target nodes of interest starting from any selected non-target nodes (acquisition path determination). The different paths can be ranked according to their probability, length, amount of flow, etc. or the combination of these values. As a consequence there is a great variety of target functions and evaluation methodologies that could be applied to this model. For example well established mathematical formulation and algorithms can be adapted from solutions of the maximum flow problem of multiple source multiple sink flow networks [3], or the algorithms mentioned in the previous chapter [2].

In addition the model can be extended with the introduction of the actual flow/capacity ratio (f/c) parameters as flow control valves applied to the edges. In this way the decision of the proliferator to change the actual flow of the processes can also be modelled (by changing the f/c value of an edge). Alternatively the similar set of parameters can be used to model the efficiency of the safeguards measures applied to a given process (f/c would be then derived from the detection probability of inspections).

3. Proliferation models

In order to use the transportation or the material flow model for acquisition path analysis, the relevant stages and processes of the nuclear fuel cycle – from the uranium mine to the weaponization of HEU or separated Pu – should be represented as a directed graph similarly to the Physical Model developed by the IAEA as illustrated in Fig. 3.

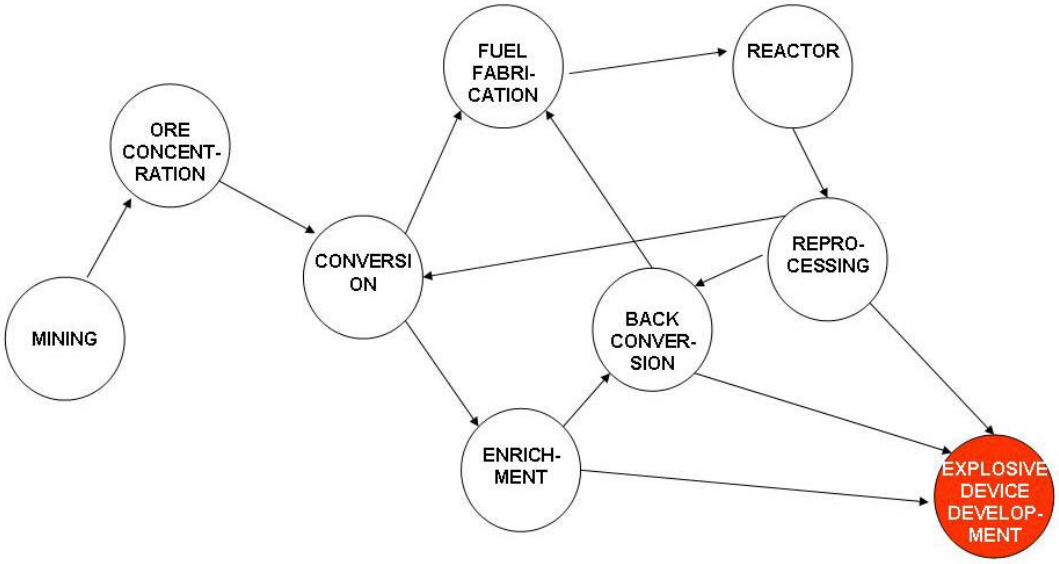

Figure 3. Directed graph illustration of the generic nuclear fuel cycle

In the following sections the application of the directed graph approach for three different levels (generic, state and facility level) are discussed in more detail.

3.1. Generic Proliferation Model

In the generic model the nodes are simply the various stages of the fuel cycle and the edges are the possible connections. For the purposes of analysis the directed graph can be described with two different types of data sets: matrix representation or adjacency list. In the matrix representation, the cell in row *i* and column *j* has the distance of node *j* from node *i* if node *i* is directly connected to node *j* and is blank otherwise (Fig. 4a). The adjacency list is a simple list of all edges ant their weights in the form (starting node - ending node; distance) (Fig. 4b).

	mining	ore concentration	conversion	enrichment	back conversion	fuel fabrication	irradiation	reprocessing	weaponization
mining		1							
ore concentration			1			1			
conversion				1		1			
enrichment					1				1
back conversion						1			1
fuel fabrication							1		
irradiation								1	
reprocessing					1				1
weaponization									

a

FROM		TO		
(mining	-	ore concentration	;	1)
(ore concentration	-	conversion	;	1)
(ore concentration	-	fuel fabrication	;	1)
(conversion	-	enrichment	;	1)
(conversion	-	fuel fabrication	;	1)
(enrichment	-	back conversion	;	1)
(enrichment	-	weaponisation	;	1)
(back conversion	-	fuel fabrication	;	1)
(back conversion	-	weaponization	;	1)
(fuel fabrication	-	irradiation	;	1)
(irradiation	-	reprocessing	;	1)
(reprocessing	-	back conversion	;	1)
(reprocessing	-	weaponization	;	1)

b

Figure 4. Matrix representation (a) and adjacency list (b) of the generic model shown in Fig. 3.

Obviously, Fig. 4 is a very simplistic description of the fuel cycle and its "analysis" leads to a trivial result. (It is like a map of Europe with only the capital cities displayed on it.) But – using the analogy of road maps – it is pretty straightforward how to make the graph of the nuclear fuel cycle richer in detail, more sophisticated, more realistic.

The first step is to look into the more detailed internal structure of the nodes. The same way as a town's internal street structure can be incorporated into a route planning application without a major effect on the town's connectivity to other towns, the various major stages of the fuel cycle can be easily enriched in detail. E.g. mining may have different forms (open pit, underground, in situ leach, uranium extraction for coal ash or sea water) or enrichment can be broken up into various technologies (diffusion, centrifuge, electromagnetic, laser, chemical). However, mining products always go to ore concentration plants and all enrichment facilities receive input from conversion. Thus the number of nodes can easily be increased since for each new node only a few new connecting edges have to be defined.

Another analogy of the roadmap that is at hand to use is that nodes and edges may have various attributes. A node can be a settlement (village, town, city), but also a museum or a filling station. An edge might be a motorway, a country road or a city street (with different speed limits) but also a ferry or railway. Also, a road can be an existing one, one under construction or only planned. Based on these attributes different views of the map can be generated with only those locations and roads that meet preselected criteria. In a similar way, various attributes can be assigned to the nodes and edges of the nuclear fuel cycle graph. For example, a node might be a material type or a process in a facility; a process might require common or high technology (diffusion vs. laser enrichment), might be industrial or in the experimental phase (traditional mining vs. uranium recovery from coal ash or sea water). Other important attributes (declared or clandestine; existing or non-existing; under safeguards or not) can also be added. In such a way, various types of analyses might be run on subsets of the graph meeting different criteria (e.g. only common technology, only declared facilities, etc.).

Finally, the key element: the edges should be assigned a set of quantitative parameters which will then be used to compute their weights and the distances for the shortest path algorithm and other types of analysis. The set of parameters may include the processing time needed to produce 1 effective kg of material, the cost of production, the production capacity or other quantitative data that might be relevant from the proliferation point of view. In general, the weight of an edge can be defined as any computable function of its parameters.

In summary, for the directed graph approach, the creation of the "generic proliferation model" would require the following elements:

- an agreed set of material types and technological processes (nodes of the fuel cycle graph) that describe the current technological possibilities relevant to nuclear weapons acquisition (including processes in experimental phase) in adequate detail (i.e. the matrix of nodes);
- the set of all possible direct routes between materials, processes (edges of the fuel cycle graph) that current technology permits (i.e. the adjacency list or the connectivity matrix);
- an agreed set of attributes to be used for the nodes and edges to facilitate the selection of appropriate subsets of the nuclear fuel cycle graph;
- an agreed set of parameters to be used for quantification of the weights of the edges;
- a set of weight functions to be used for various analyses (e.g. shortest path).

In a shorter, mathematical formulation the generic proliferation model can be fully described with the connectivity matrix $\boldsymbol{C}[nodes, nodes, attributes, parameters]$ or – equivalently – with the adjacency list $\boldsymbol{A}[from\ node – to\ node\ ;\ \{attributes_m\};\ \{parameters_n\}]$ and a set of predefined weight functions $\boldsymbol{W_{i,j}}=\{F_k(parameters_{i,j})\}$.

3.2. The Proliferation Model of a State

Generating the proliferation model of a given State from the generic model is pretty straightforward: one should simply substitute the attributes and parameters of the generic model with State specific values. These State specific values should be derived from the State evaluation process. State specific attributes describe e.g. the existence or non-existence of a given fuel cycle stage in the State

(e.g. enrichment or laser enrichment) while State specific parameters are minimum, maximum or average values for the State as a whole (total amount of material, maximum throughput of a process, minimum process time, etc.).

C= *C[nodes, nodes, attributes=State attributes, parameters=State parameters]*
A= *A[from node – to node ; {attributes=State attributes}; {parameters=State parameters}]*
$\boldsymbol{W_{i,j}}$=$\{F_k(parameters_{i,j})\}$

Several attributes and parameters have specific significance in the context of the proliferation analysis of a given State. For example, if the attribute describing the existence of a node (a given stage of the fuel cycle) *[exists=yes/no]* are allowed to take a broader set of values *[exists=yes/under construction/planned/no]* and in addition the nodes have a parameter describing the likelihood of their existence in the State *[0<=likelihood<=1; likelihood=0 if exists=no; likelihood=1 if exists=yes]* then the analysis of the existing fuel cycle can be easily amended with taking into consideration possible undeclared materials or processes.

Once the proliferation model of the State has been created (the values of all attributes and parameters has been determined) it is relatively easy to keep it up to date. Attributes need to be modified only when information about the existence of various nodes (fuel cycle stages) changes (e.g. the existence of undeclared laser enrichment). Similarly, parameters need to be re-evaluated only when total inventories of material in the State changes significantly, new facilities are commissioned or shut down, process parameters of existing ones change, or the confidence in the non-existence of undeclared materials or facilities (the likelihood of existence) change.

3.3. The Proliferation Model at the Facility Level

The proliferation model of the State as described above is still a “generic” model in the sense that it represents a summary overview of the State’s fuel cycle with general technological capabilities, total capacities etc. without providing a detailed description at the actual facility level. In this chapter we discuss the possibility of using the directed graph approach to develop a detailed realistic representation of the actual fuel cycle facilities and their relationships (connections) within a given State. For this purpose, we again use the road map analogy.

On the road map of a country, a town (node) is represented with the set of roads leading into (entry) and out (exit) of it, and the minimum or typical distances (or travel times) between any pairs of entry and exit points through the town. Similarly we can look at any fuel cycle facility as a node on the fuel cycle graph with a set of defined entry points (various types of nuclear material the facility may receive and process) and exit points (the material types the facility produces) and the set of attributes (technological possibility, capability, declared/undeclared processes) and parameters (process time, capacity, throughput) describing the connectivity of the entry and exit points within the facility. For example a nuclear power plant may receive fresh fuel (declared input) or natural or depleted uranium targets for irradiation (undeclared input) and converts them to spent fuel (declared output) and irradiated targets (undeclared output). The process time and production capacity depends on the design, the thermal power and the operating parameters of the reactor. A low enriched uranium fuel fabrication plant designed to produce LWR oxide fuel from UF_6 normally receives <5% enriched UF_6 and produces UO_2 pellets, fuel rods and assemblies, but may also receive other types of uranium input (oxide powders, >5% enrichment) and may be capable of producing other products than those declared (e.g. depleted or natural uranium targets). Also, any material received at any facility (any input) can simply be shipped out without processing, i.e. all inputs might be directly connected to some outputs. The graph representation of the above examples (“nodes”) and their possible connections in the fuel cycle are given in Fig. 5.

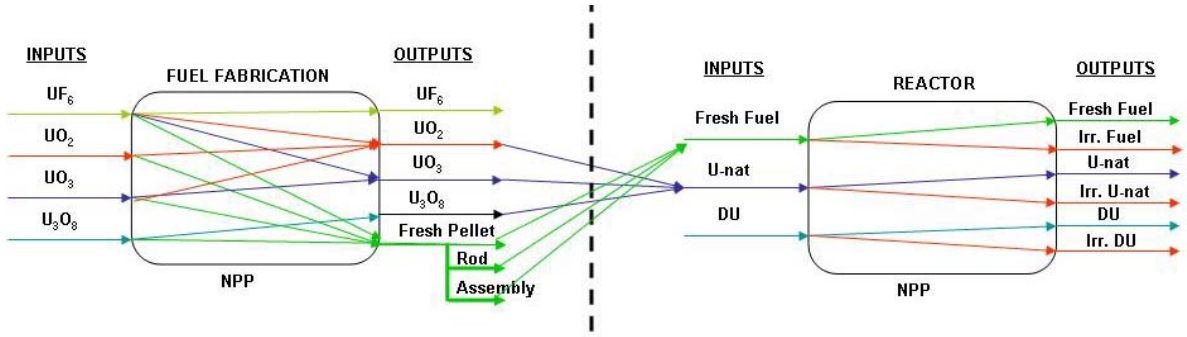

Figure 5. Graphical representation of an NPP, a fuel fabrication plant and their connectivity.

Since there are only a limited number of nuclear facility types (fuel fabrication plant, research reactor, NPP, etc.) it is easy to create their corresponding graph and the related connectivity matrices or adjacency lists and use them as building blocks to create the entire graph of the fuel cycle of a given State. If two facilities have the same list of inputs and outputs, they can be represented on the graph with the same type of building block, however, their internal structure (the values of attributes and parameters) can still be different. An example of this would be the various types of NPPs: LWR, PWR, RBMK, HFGR, etc. These categories could even be further divided (e.g. LWR: VVER-440, VVER-1000, AP-1000, etc.) and within such subgroups even the attribute and parameter values would be identical. These standardized building blocks can then simply be used to build the facility level fuel cycle graph of the State.

Since inputs and outputs are standardized, the creation of all possible connections and analysis of the resulting facility level proliferation graph can be done by appropriate software applications.

4. Creating, maintaining and analysing State models

4.1. Creation and maintenance

The model approaches described above are modular in nature, therefore by integrating the building blocks of the generic proliferation model in either model representation into the system, development of a state specific (and facility specific) proliferation models can be realised by virtual drag-and-drop methodology with using state/facility specific parameters and evaluations.

Using this modular approach would have several (crucial) benefits:

- allows a large number of items items on the graph;
- easy creation of complex graphs using simple drag-and-drop method;
- connections of nodes can be easily automated (software)
- provides for utilization of large number of existing graph analysis algorithms;
- provides for utilization of many existing graph visualization software tools;
- requires little effort to maintain the database (connectivity matrix, adjacency list): only few attributes and parameters need to be adjusted or maintained, most of them can be used with their default values.

4.2. Attributes, Parameters and Agency Safeguards Features

Whether the proposed models can be successfully used for IAEA safeguards purposes or not, depends largely on whether suitable attributes, parameters and weight functions could be defined to support the evaluation of safeguards relevant features of a State's nuclear fuel cycle. Several suggestions were already given in the previous discussions. In this final chapter we discuss a few possible options.

4.2.1. Conversion time

In Agency safeguards the “conversion time” of a given nuclear material type is defined as the time required for the production of a significant quantity of weapons grade material from the given material with the assumption that all necessary technology and facilities already exist. If the weights of the edges of the nuclear fuel cycle graph are assigned the processing time required for 1 effective kilogram of material, then the result of a shortest path analysis run for all nodes as starting points will be analogue to the “conversion time” for all nodes, i.e. all material types and fuel cycle stages. The analysis can be easily performed with or without considering undeclared facilities. Generic conversion times can be derived from the generic proliferation model, but the analysis of the State’s facility level model will result in State specific conversion times.

4.2.2. Safeguards measures

Safeguards measures such as material verification or containment can be easily incorporated in the models, through the introduction of a parameter proportional to the detection probability of the applied measure and an attribute (yes/no) indicating whether the safeguard measure is applied or not. For example in the material flow model, valves might be added to represent the blocking of diversion routes, or partial cross sections may simulate detection probabilities. Then the models can be easily analysed with or without “applying” various safeguards measures at various pipes.

4.2.3. Effectiveness

The effectiveness of a safeguards measure applied to a selected point in the fuel cycle can be defined as the increase of the distance of the selected point from the destination (nuclear weapon). The effectiveness is maximal if the measure totally disconnects the source node from the destination.

5. Summary

The IAEA has recently requested Member State support in the development of acquisition path analysis methodology and applicable software tools. To support this effort, the initial analysis of the applicability of the directed graph methodology and corresponding network modelling software solutions were carried out. In this paper we described the two possible representations of the nuclear fuel cycle as a directed graph: the transportation model and the material flow network model. It was shown that both models are suitable to form a general framework for the acquisition path analysis methodology. They could be described with similar mathematical formulas and solved with similar analytical methods, which can easily be built into existing software environments readily available to build, manage and visualize large network modelling problems.

If the concept outlined in this paper finds wider acceptance in the professional community, the key tasks of developing acquisition path analysis methodology for IAEA safeguards purposes would consist of the following steps:

- the determination (in appropriate detail) of the set of material types and technological processes that describe the current technological possibilities relevant to nuclear weapons acquisition;
- the identification of all possible (relevant) direct routes between materials, processes that current technology permits;
- the identification and evaluation of the attributes and parameters to be used for the nodes and edges of the graph models;
- the definition of the required analytical goals (weight functions) and corresponding mathematical algorithms;
- the selection (and customization, if needed) the most suitable graph modelling and data management software environment.

We are confident that even if the validity in the real world situations of the actual absolute values obtained through any graph analytical methods might be questioned, the proposed approach would

definitely provide a sound objective basis for the comparison of different safeguards measures, or comparison of different States.

Acknowledgement

The authors express their thanks to Mr Akos Peto (IAEA) for initiating this topic and for his interest in and valuable contribution to this work.

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Evolution of civilian plutonium inventories for different fuel cycle strategies in plutonium handling countries

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Abstract:

The global stockpile of separated civilian plutonium has reached about 250 tons heavy metal (tHM) in 2009 and is now almost equal to the amount intended for military purpose. Most of the civilian material is owned by Nuclear Weapon States plus Japan, but other states have also expressed their will to start commercial scale reprocessing and recycling in the future. As separated plutonium is a direct-use material and bears major proliferation concerns, it is important to know how these inventories might evolve in the future. By taking country specific scenarios of nuclear energy and fuel cycle developments into account, calculations of these inventories up to 2050 are performed with the "Nuclear Fuel Cycle Simulation System (NFCSS)" of the International Atomic Energy Agency (IAEA). Results show that stocks of separated plutonium will increase significantly in the next decades as a result of nuclear energy development and planned fuel cycle back-end operations. A doubling of the current amount until 2050 seems possible. Even if reprocessing was slowly abandoned and recycling in mixed-oxide (MOX) fuel continued on today's level, separated plutonium stocks would exceed current inventories by far in 2050.

Keywords: plutonium, Nuclear Fuel Cycle Simulation System, nuclear fuel cycle

1. Introduction

It is generally agreed that the production of fissile material is the most difficult, time-consuming and expensive step on the way to a nuclear weapon. Besides highly enriched uranium, plutonium is qualified for this purpose and has been intensively produced by today's nuclear weapon states. Plutonium does not occur naturally and can only be generated by neutron capture of uranium in nuclear reactors. This process can be optimized for weapons material production, but happens also inevitably in every civilian nuclear reactor to a certain extend. At the beginning of 2010, the estimated worldwide amount of separated plutonium, which can be directly used for nuclear weapons, was about 485 tHM possessed essentially by eleven states [1]. This stockpile is shared almost equally between the civilian and the military sector. The latter will prospectively decrease in the future: Although India and Pakistan continue to produce and separate plutonium, the two countries with the largest stocks, Russia and the USA, have begun to implement disposition programs to reduce their material (together they are responsible for ca. 92% of all military stocks). On the other hand, separated civilian plutonium is thought to increase because of on-going or beginning reprocessing activities in France, UK, Russia, Japan, China and India besides of a few countries with minor activities. The civilian production, separation and use of plutonium constitute major proliferation concerns because it can also be utilized as fissile material in nuclear weapons [2]. It can be assumed that a possible nuclear renaissance, especially encouraged by the wish of climate change mitigation, would increase civilian stocks significantly in the future making international safeguards more complex and expensive.
The aim of this work is to analyse and forecast the prospective development of civilian plutonium inventories. For this purpose national nuclear fuel cycles are modelled with the "Nuclear Fuel Cycle Simulation System (NFCSS)" of the IAEA. As far as data exist, the historical development of civilian

plutonium stocks (separated and not separated) can be compared with the model results. Country specific scenarios that take future capacity developments and fuel cycle back-end strategies into account are applied to calculate amounts of separated plutonium as well as plutonium contained in spent fuel up to 2050 produced by thermal reactors. For this purpose, key parameters regarding nuclear energy usage and fuel cycle have to be determined closely by taking existing plans of stakeholders into account.
Although international cooperation takes place, the important policies and strategies for handling plutonium are national ones. Considering this and taking into account that countries pursue different policy strategies for handling civilian plutonium, the calculations are conducted on a national basis for the most important countries that handle separated plutonium. The fuel cycles of countries which fulfil the following criteria are modelled:

1. Civilian reprocessing in the present or concrete plans to do so in the future or

2. MOX usage in the present or concrete plans to do so in the future.

In both cases, nations are compelled to handle significant amounts of separated or at least unirradiated (in form of MOX) civilian plutonium. These criteria are fulfilled by the following countries: China, France, Germany, Japan, India, Russia, UK and USA.[1] This list of countries simultaneously represents most of the largest nuclear energy users in the world. Together, they are responsible for about 70% of the global installed nuclear capacity. Most of these countries publish annual information about their current stocks through the “Information Circular 549” [3]. Although these declarations possess only limited accuracy and completeness, it is at least possible to compare them with the modelled results. It is important to note that breeder reactors will not be included in the subsequent models and analyses. At the moment there is no indication for the conclusion that breeder reactors will become relevant within the next decades. In fact, their economic handicap compared to the once through cycle is unlikely to vanish before the end of the century [4]. Even for those scenarios of the International Panel on Climate Change (IPCC) that assume the largest increase in nuclear energy, enough uranium ressources are available at sufficiently low price for plutonium not to be required as fuel by the end of 2100 [5].

2. Model and basic input parameters

NFCSS was developed by IAEA in 1996. In its current form, NFCSS is a web-based software, which is available for authorized users of the IAEA member states. The program is able to simulate a complete fuel cycle on the basis of annual time steps and to calculate its associated material flows as well as the changes in fuel compositions by depletion. It is possible to create different energy scenarios for one reactor unit, a whole reactor park or a full nuclear fuel cycle [6].
Figure 1 gives an overview of the working scheme of the computer code. The calculation mode of NFCSS consists of two modules: The module named “CAIN” (Calculation of Actinide INventory) is responsible for calculating the actinide inventory which changes with irradiation time in the nuclear reactor. The code works with one-group cross sections and includes libraries for standard reactor types. Cross sections, specific power of the fuel and neutron flux are provided as reactor specific, averaged values over the whole burn-up. The annual material balance is calculated by “VISTA”, which is the other module responsible for material flow calculations. The generated output lists a whole set of parameters, beginning with the natural uranium and fuel production requirements. For the goals of this work, the spent fuel discharge and its isotopic composition are of special interest, as well as the material involved in reprocessing and MOX usage.
The most important input parameters are nuclear capacity, the types of reactors, their annual load factors, average enrichment, discharge burn-up, reprocessing capacity, MOX-share, time lag between certain processes and loss coefficients. For the purpose of this work, two types of parameters are differentiated: Predetermined parameters and scenario parameters. The first group contains physical or technical parameters that will not be variegated in the scenario based calculations, e.g. thermal efficiency, fuel residence time, burn-up etc. These values are not necessarily fixed over time or free of presumptions but generally independent from political decisions. In contrast, the scenario parameters are deliberately changed according to scenarios calculated. They include capacity development and reprocessing and recycling strategies, which are based primarily on political decisions to less extent

[1] Belgium, The Netherlands and Switzerland have also a minor MOX program. These countries are not considered here.

on technical aspects. Scenario parameters have major influence on the model and underlie the most uncertain future development. All values have to be specified for every modelled year. For historic development, the values for both parameter types are not always known. Various references are used to compile as much information as possible for providing an accurate input.

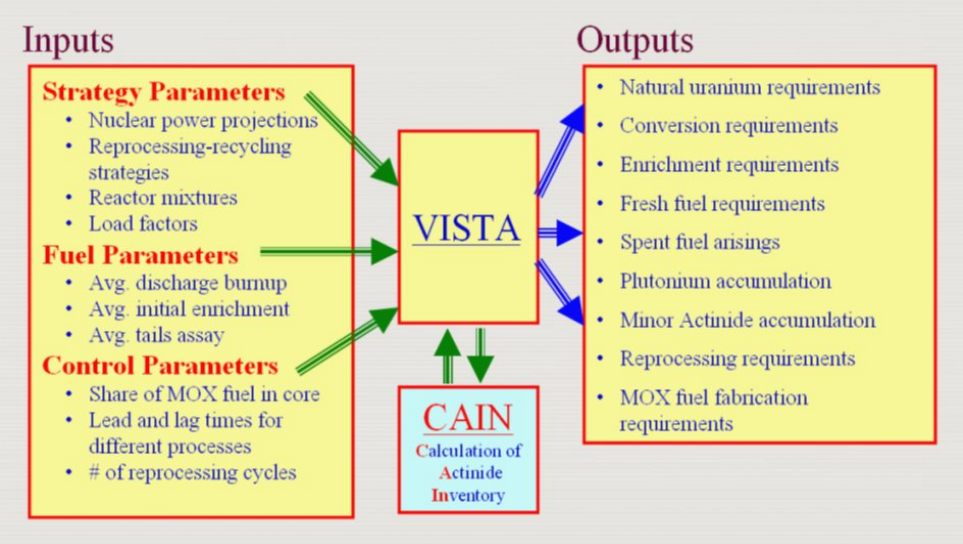

Figure 1: Working scheme of NFCSS (taken from [2])

2.1. Predetermined input parameters

Predetermined parameters are not changed in different scenarios in order to keep them comparable to each other. These are: load factor [%], thermal efficiency [%], tails assay [%], fuel residence time [a], initial enrichment [weight-% ^{235}U], initial enrichment fuel type 2 [weight-% Pu], discharge burn-up [MWd/kg], loss factors (reprocessing, fuel fabrication), cooling times and processing times. For information on historical development, databases play a key role. The “Power Reactor Information System” (PRIS) of IAEA provides historical information on the installed capacity and annual load factors for every reactor type in a country [7]. The future load factor for 2011 - 2050 is estimated by calculating the reactor type and country specific average over 2001 - 2010. Thermal efficiency is a thermo-dynamic characteristic and essentially a constant for a reactor type. Fuel residence time depends on reactor type and core management, but does not change significantly with time if operation follows economic incentives [8]. Table 1 shows the standard values of these parameters for every considered reactor type.
The actual historical and expectable future evolution of burn-up is important information for the model input and also determine initial fuel enrichment. The increase of burn-up over time probably varies from plant to plant very much. So, only averaged values for a reactor type can expected to be useful for the simulations examined in this work. A few sources provide these types of information, partly differentiated between Asia, West-Europe and the USA [9, 10]. With help of these references, a data set of averaged burn-up values as function of time has been compiled. As the simulations are intended to cover a period up to the year 2050, simple extrapolations with constant gradients are made for future burn-up development.

2.2. Scenario parameter

Scenario parameter include: installed nuclear power (reactor type specific) [MWe], fuel type 2 share (MOX) [%], reprocessing ratio [%] and reprocessing capacity [tHM]. As the aim of this work is to assess country-specific scenarios, energy and fuel cycle prognosis are consequently also needed for every country. The following references are consulted for defining the scenarios used for the simulations:

- WEO – 450: Scenario of the World Energy Outlook in which energy production consistent with the aim to keep the increase of global average temperature below 2^{0}C, or 450 parts per million of carbon-dioxide equivalent [11].

- WEO – New Policies: Includes recent national commitments to reduce greenhouse gas emissions (“Copenhagen Accord”) and to address other environmental and energy-security concerns [11].

- NEA: For countries that are not explicitly mentioned in the WEO scenarios (WEO gives for example only aggregated data for Europe as a whole), the scenarios of the forecast of the Nuclear Energy Agency (NEA) is used [12].

- Prognos: In 2009, Prognos published a study on behalf of the German Federal Office for Radiation Protection, in which projections for the future global development of nuclear energy is analysed critically [13]. It takes possible barriers for building new plants like lacking infrastructure, fuels supply, financing, political stability and generates a numerical value for the likelihood of realisation of new nuclear power plants. This information is combined with the WEO - Current Policy scenario to develope a conservative prognosis of nuclear energy development. The name for scenario as used in the subsequent case studies is “Prognos”.

In order to present a convenient overview that is not too complex, the country-specific fuel cycle back end strategies are not variegated here but only reflect current political goals. They are derived from the *country briefings* of WNA [14].

3. Results

In the following subsections, the results for world inventories as a whole as well as case studies of two analysed countries are presented. Modelling the world as a whole allows to easily study the implications of major changes in back-end strategies. France and Japan are chosen for case studies, which are meant to exemplify the calculations done for every of the considered plutonium handling countries. For the two cases, a short introduction to the present situation, history and political goals for the future regarding nuclear energy and fuel cycle is given. Finally the results for all considered countries are presented and discussed.

3.1. World

Before presenting the detailed case studies of countries that are especially interesting regarding back-end fuel cycle operations, the plutonium inventories on global scale are analysed. Historical information on load factor and capacity development is taken from PRIS. Past reprocessing activities and MOX usage were modelled with the help of various resources like [15] and the WNA country briefings [14]. In addition it is assumed that all Magnox fuel is reprocessed. MOX share is assumed to be the same for PWR and BWR and only once-through plutonium recycling is assumed. The reactor type share for future added capacity is chosen equal to the share as in 2010 but taking shutdown of all RBMK, AGR and Magnox after the end of their lifetime into account. Three reprocessing and recycling scenarios are included in the following NFCSS calculations: continuation of current operations (REP medium - MOX low), phase out of reprocessing during 2020 and 2040 (REP low - MOX low) and duplication of current reprocessing and MOX usage during 2020 and 2040, respectively (REP high - MOX high).

Figure 2 shows the global cumulative amount of separated plutonium from 1955 to 2050 for the three back-end scenarios defined above. As scenario for the nuclear energy development, Prognos is chosen. The comparison with INFCIRC/549 declarations shows good agreement with the obtained results although it becomes worse after the year 2000. In 2009, the discrepancy is 37.5 tHM or 13%. This relatively large deviance could be caused by insufficient assumptions regarding the global MOX share which is assumed to be 3% in 2010. Due to lack of information, this is only a very rough estimation. The flattened increase after 2005 is due to THORP's shut-down. The subsequent increase is caused by its reopening and Rokkasho’s delayed start-up in 2013. As expected, major differences occur for the considered reprocessing and recycling scenarios. Doubled reprocessing activities and MOX share (6%) results in an amount of 1,210 tHM separated plutonium in 2050. Also the continuation of current fuel cycle back-end activities results in a monotonously rising inventory (880 tHM in 2050). Only the scenario with phasing out of reprocessing and continued MOX share shows a decreasing inventory with the maximum of ca. 390 tHM reached in 2031.

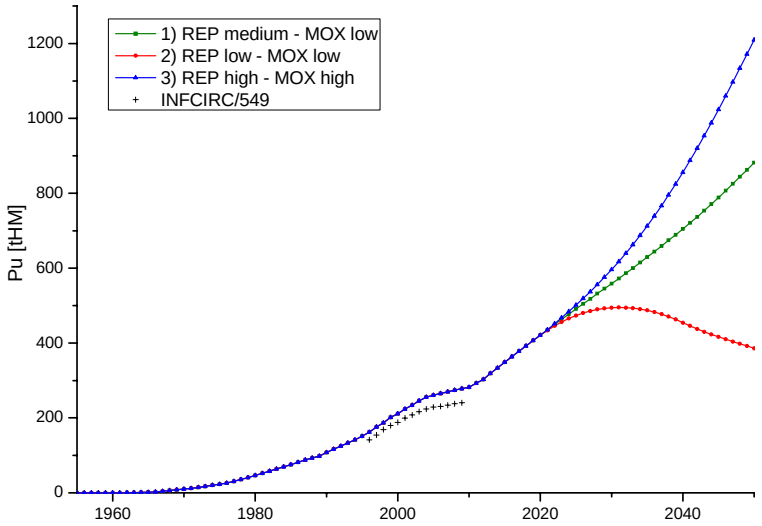

Figure 2: Global separated plutonium amount for the Prognos scenario and the three different back-end scenarios: 1) REP medium - MOX low (continuation of current operations), 2) REP low - MOX low (phase out of reprocessing during 2020 and 2040) and 3) REP high - MOX high (duplication of current reprocessing and MOX usage during 2020 and 2040, respectively).

The plutonium which is not separated is contained in spent fuel and shown in figure 3. In contrast to the separated amounts, this inventory is predominantly depending on development of energy scenarios whereas a small change in MOX share has only very little influence. According to the simulation, there are 2,070 tHM irradiated plutonium present in 2010. If reprocessing and MOX share will be hold constant during 2010 - 2050, this amount will rise to 6,580 tHM (WEO - 450), 5,695 tHM (WEO - New Policies) or 4,754 tHM (Prognos). The production from present reactors plus plants under construction in 2011 leads to an amount of 3,916 tHM in 2050.

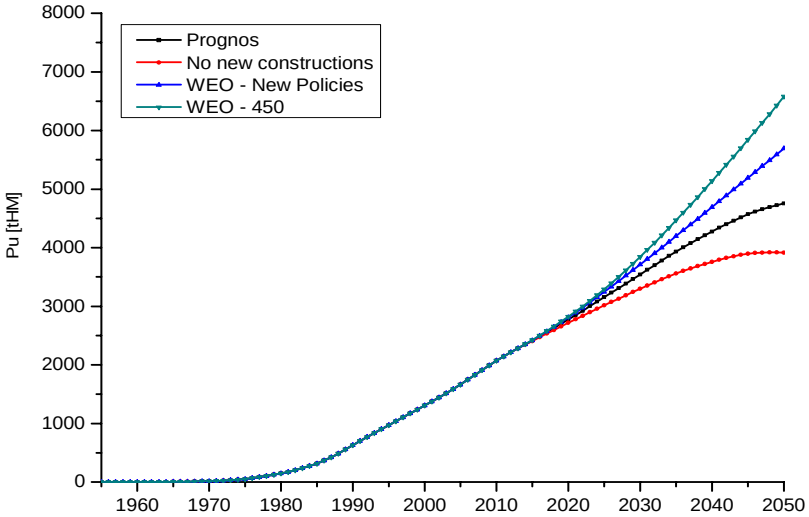

Figure 3: Global amount of plutonium contained in spent fuel according to the input scenario REP medium - MOX low (continuation of current operations).

3.2. Case study 1: France

With 59 nuclear reactors, France runs the largest nuclear fleet in Europe and the second largest in the world. All of the active plants are pressurized water reactors, although GCRs similar to the British MAGNOX-type had also been used until 1993. France reprocesses most of its spent fuel at two facilities at La Hague, UP2 and UP3, with a combined design throughput capacity of currently 1700 tHM/a. Due to this enormous reprocessing activities (both for domestic and foreign utilities), the

country has accumulated a separated plutonium stock of about 55.9 tHM. Plutonium is fabricated in MOX fuel (currently at the 195 tHM/a MELOX plant) and has been recycled in thermal reactors since 1987. In 2010, 20 reactors were using MOX fuel, with further four plants waiting to be licensed [14].

WEO scenarios cannot be used for France because it provides only values for Europe as a whole, but NEA data should be comparable to WEO - New Policies as it includes newest governmental ambitions. It is assumed for all scenarios that MOX-usage will be expanded to the last four 910 MWe-reactor units and will then maintain a constant capacity share.

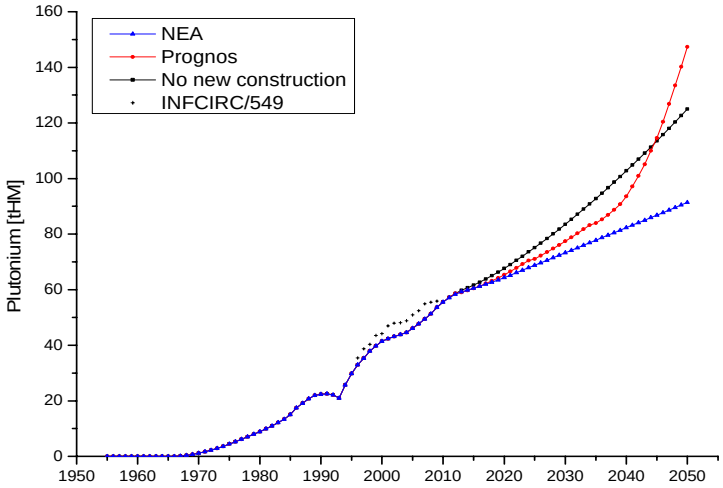

Figure 4: Separated plutonium from French nuclear reactors.

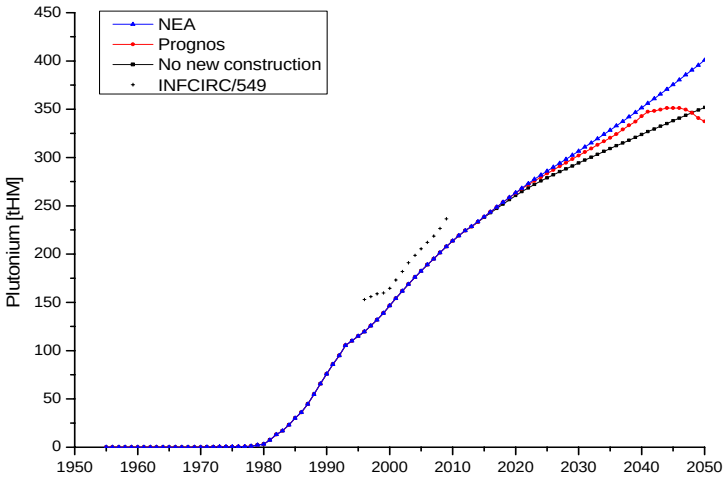

Figure 5: French plutonium contained in spent fuel.

Results for the evolution of inventories with separated and irradiated plutonium are plotted in figure 4 and 5. France declares its inventories to IAEA which means that the modelled results can be compared with this data. The local maximum of 22.5 tHM separated plutonium in 1991 is caused by combination of phase out from GCR reprocessing and shortly after this the increase of separation capacities for PWR fuel. Due to the lack of information, this development could be unrealistic, but unfortunately the INFCIRC-declarations began not before 1996 thus preventing comparison of model and declaration. From 1996 onwards, the trends of declaration and modelled plutonium amount corresponds well with an average discrepancy of 3.8 tHM in the period of 1996 - 2009. After 2010 it can be seen that the stock of separated plutonium increases in all scenarios. It reaches 91.4 tHM (NEA), 125.0 tHM (No new construction) and 147.3 tHM (Prognos) in 2050. The increasing capacity in the first case with a fixed percentile MOX-share causes a higher plutonium consumption rate and therefore the slowest growing plutonium inventory. The amount of irradiated plutonium reaches about

400 tHM in 2050 in the NEA-Scenario. Only in the Prognos scenario, more plutonium is used than produced near the end of the modelled period in 2045.

3.3. Case study 2: Japan

Japan began in 1966 with commercial nuclear energy production and expanded its reactor fleet continuously to meet the countries high energy demands. Today, 54 units and several fuel cycle stages including two reprocessing plants are operational. Start of commercial operation at the Rokkasho reprocessing plant is scheduled for the end of 2012. The plutonium is designated for MOX production and usage in LWR. The plans faced long delays, so that the first MOX assemblies had not been deployed before the end of 2009.

The country's current unirradiated plutonium stocks amount to 46.34 tHM. Most of it (38 tHM) is stored in separated form in France and the UK, where a part of Japans spent fuel has been reprocessed. Japan does not declare how much of the plutonium in spent fuel that was send overseas is still waiting for reprocessing. It can only be stated that at least the predominant part is reprocessed by now. In the following, the amount of 46.34 tHM is assumed to be Japans full stock and any more plutonium is only resulting from domestic reprocessing. The Rokkasho reprocessing capacity of 800 tHM/a is included in the calculations from 2013 onwards, the total MOX share is assumed to rise to five per cent until 2014.

Figure 6 shows how much separated plutonium Japan is going to have in the different scenarios. The amount seems to decrease only for the WEO – 450 scenario in which more plutonium is consumed than reprocessed after 2023. In 2050, the stocks would reach a value between 131 tHM (Prognos) and 16 tHM (WEO - 450).

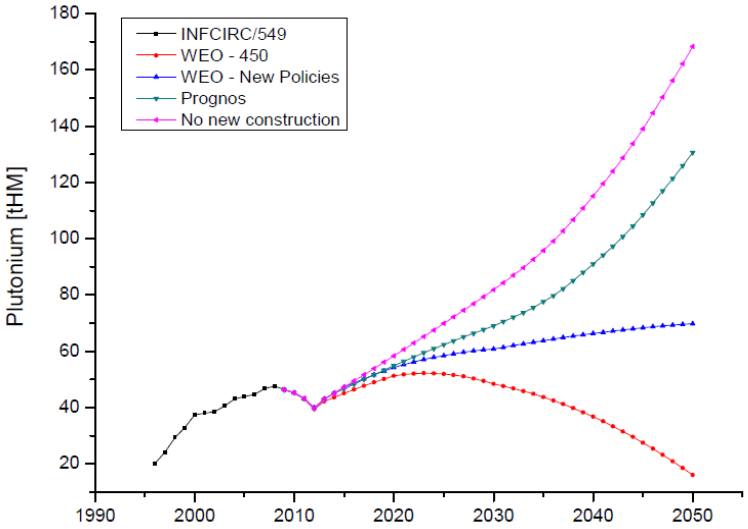

Figure 6: Japanese separated plutonium with a MOX share of 10%.

Figure 7 gives the plutonium stock in spent fuel, which will rise also. The maximum differences between the scenarios in 2050 are only around 25 tHM, with the largest amount occurring in WEO – 450. The comparison with INIFCIRC/549 shows good results. The gradient and the absolute values of the declared amounts are reproduced well by the model.

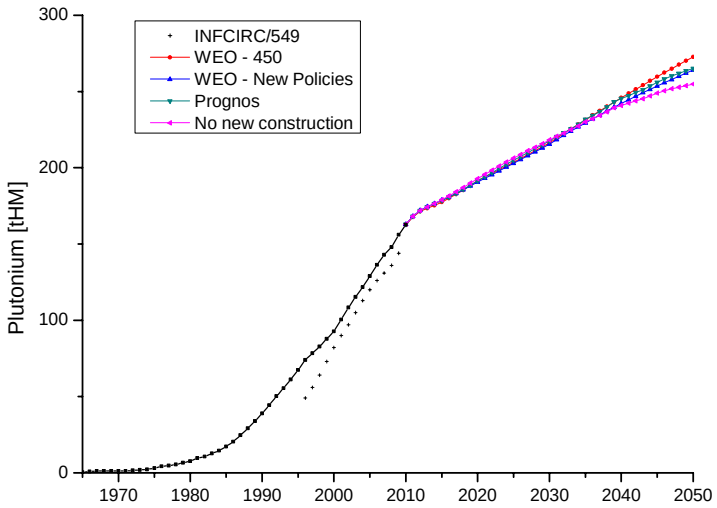

Figure 7: Japanese stocks of irradiated plutonium. From 2000 – 2010, results of the model and the declarations fit well. The outcome of the scenarios shows only little differences.

3.4 Summary

Table 1 shows the results for all modelled cases for 2030 and 2050. Different energy scenarios are assumed, whereas fuel cycle back-end parameters are chosen following official plans.

Country	Energy Scenario	Separated plutonium [tHM]		Plutonium in spent fuel [tHM]		Comments
		2030	2050	2030	2050	
USA	WEO - 450	18	0	1128	1684	Plutonium disposition program (MOX-share of 1.7%) , no reprocessing
	Prognos	18	0	1152	1598	
France	NEA	73	91	401	307	Current reprocessing capacity and MOX-share (8.6%) continued
	Prognos	77	147	337	302	
Japan	WEO - 450	48	16	216	273	Reprocessing as officially planned, MOX-share of 10%
	Prognos	69	131	217	265	
Germany	-	0	0	178	191	Future nuclear energy production capped by law
Russia	WEO - 450	17	90	258	195	Reprocessing as officially planned, MOX-share of 10% for VVER-1000
	Prognos	22	150	259	172	
UK	Prognos	84	54	81	154	Reprocessing phase out as officially planned, 5% MOX-share
China	WEO - 450	7	0	227	744	Reprocessing as officially planned, MOX-share of 5%
	Prognos	37	90	142	203	
India	WEO - 450	18	30	102	436	PHWR reprocessing (300 tHM), no MOX
	Prognos	18	30	70	161	
Total		265 – 325	281 - 602	2436 - 2591	3046 - 3984	

Table 1: Total amounts of separated and unseparated plutonium for all plutonium handling countries according to the current plans for reprocessing and recycling

4. Conclusions

The goal of this work was to derive conclusions on future implications resulting from current plans for nuclear energy usage and fuel cycle back-end strategies. Simulations of the most significant civilian plutonium handling countries taking different scenarios into account were performed. According to the results, growing separated plutonium stocks must be expected in the future: They will more than double until 2050 if current plans of reprocessing and recycling are realised. Global stocks of plutonium in spent fuel will reach about 3,000 – 4,000 tHM depending on the nuclear capacity development.

Naturally, calculations like the ones performed for this analysis give only forecasts with relative limited validity. If a few countries do not implement some key elements of their nuclear plans or if new plutonium consuming facilities (burner or breeder reactors) are put into operation, many results presented here will have to be revised. Nevertheless it was shown here that consequences of current national policies lead to a significantly increasing stock of direct-use material that constitutes rising proliferation threats. Since there is no foreseeable use for the surplus of separated plutonium the policies for nuclear fuel reprocessing should be carefully revisited.

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Item Tagging Considerations under a Verified Warhead Dismantlement Regime

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Abstract:

The AWE Arms Control Verification Research team is engaged in a program of exercises and research focusing on equipment and procedures for the verification of nuclear weapons dismantlement and storage, under hypothetical future arms control regimes.

The current range of high security Tamper Indicating Devices used throughout international Safeguards may not be suitable for use in operating nuclear weapons facilities, under the conditions imposed during an internationally Verified Warhead Dismantlement Regime (VWDR). The security restrictions of Host facilities, combined with stringent Authentication requirements, create a need for high security tagging and sealing systems and procedures that will satisfy the competing interests of all the parties involved. This paper discusses the potential implications of arms control verification for tagging system requirements, and introduces some concepts that may help satisfy those requirements.

Keywords: Arms Control; Certification; Containment; Surveillance

Introduction

The conditions under which international verification of nuclear warhead dismantlement could be conducted will be significantly different from the conditions experienced under established inspection regimes. Severe restrictions, which do not feature in current regimes, will likely be placed upon all technologies used, and could render unsuitable some technologies developed for less stringent environments. At the very least, equipment Authentication methods and operational methodologies will need to be revised and assessed for suitability, vulnerability and validity in the verified warhead dismantlement paradigm.

The focus of this paper is on the restrictions that could be placed upon containment and surveillance equipment, used in a Verified Warhead Dismantlement Regime to help maintain a Chain of Custody over Treaty Accountable Items (TAIs) [1]; and on design considerations for tagging systems that could meet those restrictions.

The Chain of Custody

The availability of information through Non Destructive Assay (NDA) of the TAI will be greatly curtailed in a Verified Warhead Dismantlement Regime, limiting the utility of NDA for fissile material accountancy through dynamic dismantlement processes (See Appendix 1). Containment and Surveillance (C&S) technologies may therefore be relied upon to a great extent in order to maintain a ‘Chain of Custody’ over the TAI, in order to establish a level of accounting over the total amount of fissile material arising from a dismantlement campaign.

The aims of a Chain of Custody are to verify that individual items are presented for and undergo dismantlement, so that no TAI can be recycled through the process; and that *all* the treaty accountable

fissile material present in a TAI before dismantlement is present afterwards, i.e. to deter and detect diversion of material.

To achieve these aims, all Treaty Accountable Items need to be uniquely identified and contained within a monitored boundary at all times, through which no material can pass without raising the awareness of the treaty-mandated Inspectorate. The use of containment and surveillance technologies of various kinds could aid the process, but the Inspectorate must also maintain trust in the function of such technologies. Inspection C&S equipment may therefore also require unique identification and protection from tampering.

This paper will briefly outline possible restrictions in Verified Warhead Dismantlement Regimes and then track their effects on the 'simple' task of tagging an item with a unique identity, and then verifying that tag. The example given discusses tagging concepts that use a random distribution of particulate, such the Reflective Particle Tag developed at Sandia National Laboratory [2,3]. The paper will show that Regime restrictions can create the requirement for the tagging system to exhibit new capabilities and functions, which must be accounted for during the design stage of system development. Some options and operational concepts are introduced to suggest how suitable tagging systems might meet the requirements imposed.

The thought process, and similar requirements, could well apply to all technologies deployed in the course of a VWDR. The design process for suitable C&S systems must account for such restrictive conditions, in order to develop suitable surveillance and containment solutions for warhead dismantlement verification.

A Model Verified (Nuclear) Warhead Dismantlement Regime Scenario

The model VWDR presented below is intended to reflect a regime as it might apply from the point at which Inspectors first encounter a TAI to the point at which all classified characteristics are erased from the fissile material arising from the disassembly process. Once classified characteristics are eliminated from the material, standard Safeguards parameters might apply.

The Model

Within the Model, the potential threat, as perceived by the Inspecting Party, is similar to that in a Safeguards regime: A threat constituting the entire state itself, able to employ all necessary resources in order to defeat a monitoring system emplaced within its own facility. If the decision is taken, the Host could spend unlimited amounts of time devising ways to defeat the system.

The aim of the Verification Regime is to confirm that all fissile material from a dismantled weapon system enters into a monitored storage facility (eventually, perhaps, as unclassified material under more traditional Safeguards), by deterring the Host from spoofing, replacing or diverting its own material, or from otherwise 'cheating' the regime by retaining weapons grade materials through subterfuge. A successful inspection regime will therefore need to be able to detect and provide evidence of an attempt to undermine it.

The overall objective of the Regime, as well as the threat, is therefore analogous to that of a Safeguards regime. However, in this Model, the warhead dismantlement verification environment diverges from current regimes on four fronts:

- Host security concerns dominate the inspection regime. A warhead disassembly complex contains highly sensitive national security information, which the Host must protect. The need to protect information does not cease with the decision to reduce stockpile numbers;
- Non-proliferation concerns affect decision making. The Model Host State is a Nuclear Weapon State as defined by the NPT and so has NPT Article I commitments to uphold. The Model Inspectorate also has NPT Article I/II commitments to consider.
- Health and safety considerations need to account for the presence of explosives;
- The dismantlement process is dynamic, but the ability to halt or delay dismantlement operations once the process begins is limited.

These differences encapsulate a paradigm shift away from other regimes and the shift affects the selection and use of technology to be deployed within the Scenario. The competing interests of the involved Parties gives rise to the fact that all inspection equipment, and its method of operation, must satisfy competing demands of both Host and Inspector.

In this Model, technologies deployed by the treaty-mandated Inspectorate during warhead dismantlement cannot be allowed to monitor security sensitive operations or collect classified information. Information relating to Warhead design, facility operations and physical security of the warheads is all treated as classified.

The conditions of the Model are now explored to see how they could affect the operation of container and surveillance, and particularly tagging, systems, and the designs of those systems.

Certification Requirements in the Model Regime

The Host Party has the right and necessity to protect large amounts of sensitive information. The Host must therefore certify all equipment as suitable for use within its facility, to ensure its own interests are satisfied. Certification measures will account for Host security, non-proliferation and safety concerns.

The Certification process will include measures undertaken by the Host to ensure all equipment functions only as agreed and designed and remains in a pristine state; to ensure that, if deployed within its agreed capacity, the certified item cannot collect or transmit any data not explicitly sanctioned for release by the Host; whether or not operated in the agreed manner. The Host will therefore require unrestricted, sole access to all equipment in order to certify it.

Certification will have to consider the manner in which the equipment is deployed and the mode of operation. If certain technologies need never be deployed into sensitive areas, the Certification process might be easier. Likewise, data emanating from versatile equipment may be released more simply if that equipment has been operated under conditions which constrain the type of data that could have been collected. For instance, Inspector use of a camera could result in loss of sensitive information, but if operated by Host personnel, the data collected could be controlled and might therefore be considered suitable for release.

Equipment required to enter sensitive facilities within a disassembly complex may not be certified by class or type; but each individual unit may need certifying, though the author suggests that bulk raw materials could be sampled. Once certified, the Host must ensure Inspector access to equipment takes place under controlled conditions to ensure the equipment remains in a pristine state. If the Host loses custody over equipment it may have to be re-certified before being allowed into a sensitive facility. Any equipment that cannot be certified by the Host will not be allowed within the facility.

Within this Model Scenario Inspection equipment that is never required to enter a sensitive facility may be certified for use in on weapons complex, but the arrangements must be put in place to ensure such equipment cannot communicate with any technologies destined for sensitive facilities

The requirement for Host Certification of all Containment and Surveillance systems generates the need to re-evaluate existing systems because equipment design; equipment provision; Inspectorate Authentication, inspection and data analysis methods could all be affected.

Certification Consequences in the Model

Requirements for Standardised Equipment Designs

In order to satisfy Certification needs, both parties must agree a standard design for every piece of equipment used in sensitive facilities. Any unexpected deviations for the agreed standard could lead to a rejection of that piece of equipment, at least until it is proven to be inconsequential to either of the Parties. The consequence of this is that there can be no hidden functionality and no hidden features or design details in any equipment used within sensitive facilities during verification of warhead dismantlement. There may be a requirement for full design disclosure of any commercially available systems or any equipment already in existence.

Tags and other Containment and Surveillance systems should not rely on covert technology to ensure their integrity, detect tampering, or defend against counterfeiting in this situation. Subtle or complex markers are not sufficiently robust to use in an environment where the Host holds all design details. In relation to tagging requirements and identification of inspection equipment, identifiers have to be unique and randomly generated.

Host Supply of Equipment

Rigorous Certification measures, combined with Full Disclosure of all equipment designs, may result in all equipment effectively becoming 'Host Supplied' [4]. Host supply of equipment creates a situation akin to that of 'joint use' equipment in Safeguards. This is very likely the case for any systems required to enter sensitive facilities within a disassembly complex, within this Model Regime.

Separating systems into subsystems designated for 'sensitive' or 'non-sensitive' facilities may offer a way to reduce the Certification requirements, but the tagging system architecture must be designed for this. A data diode should be incorporated into such architecture, thus ensuring information can only flow *from* the authorised collection device *to* the analysis equipment [5]. Operating procedures incorporating this must also be clearly defined in order to be considered for Certification.

Technologies that the Inspectorate can come to trust, even if provided by the Host, are important in this Model Regime. Complexity of operation can make the task of Authentication more difficult and so 'simple' technologies, as described earlier, are preferable [6].

In an environment where the Host provides equipment, counterfeiting becomes a serious concern. Intrinsic features of a material or technology might be attractive in other regimes for proving individuality or uniqueness, but if the Host supplies that intrinsic signature, it may be hard to determine whether something is truly unique or an excellently made 'pseudo-signature', counterfeited multiple times before a single copy is given to the Inspectorate for use.

In relation to physical tagging techniques, intrinsic tags, or those made through random processes, cannot be certified and authenticated unless the creation method is agreed and verified by all Parties, and the tags remain under dual custody from the point of manufacture.

If the Inspecting party manufactures the tags, individual tags may require (destructive) Certification testing to ensure they are all meet the standard design, with no additional functionality. If they are pre-made by the Host, uniqueness cannot be guaranteed. In a verified warhead dismantlement regime, technologies may benefit from a tag that is generated and applied by the Inspecting Party using certified raw materials provided by the Host, in a suitable facility under dual custody.

Generation of digital signatures by the Inspectorate may prove extremely complex if the processor chosen to perform the signing function is Host supplied, along with any other required hardware. The Inspectorate must then ensure that the cryptographic private key data is genuinely and privately generated in the agreed manor, and remains secure at all times. Authenticating a device that could delete malicious code if interrogated, but which otherwise could record or introduce key information at a time designated by the Host is a significant challenge.

Careful consideration will need to be given to whether a system could be certified if it involved use of Inspectorate provided hardware for digital data signing. If, the data collection device were jointly designed with no additional functions or sensors; the system was operated by the Host, and the operation connecting the signing hardware with the camera were performed under secure conditions away from sensitive facilities, maybe the system could be configured in such a way that it could not collect or deliver any information other than intended, even if given instructions to do so by the Inspector provide hardware. The author suggests that the entire operating system and procedure, including use of Inspector provided hardware, might be considered for Certification in such a case, but would require much more detailed analysis concerning feasibility and vulnerabilities.

Environmental Isolation and Post Deployment Equipment Retrieval

Within this model, it is likely that once equipment has entered into a sensitive facility it would not be released into sole Inspector custody unless it is first sanitised, or it could otherwise be demonstrated

that the item concerned had been effectively isolated from the environment into which it was introduced, at all times. Isolation methods would have to be demonstrated as part of the Certification process and would have to form part of the overall operational procedure for the item.

This affects the options for Inspector Authentication of equipment and for detailed analysis of hardware and data for inspection purposes, and should be a factor considered during the design stage of all containment and surveillance equipment; if retrieval is important as a matter of operation and inspection. If retrieval of certain components is not important, modular, 'Sacrificial' components may offer a solution. A system with a 'Sacrificial' component would be designed so that the designated component would be left in Host custody when no longer required.

Technologies designed to be isolated from the disassembly environment, possibly within a protective atmosphere, a physical barrier or jacket, could provide more flexibility during deployment and retrieval. The jacket would effectively become the 'sacrificial' component, allowing the core components of the system to be retrieved. Such a design would have to demonstrate that the barrier or jacket offered complete isolation. In some circumstances, a procedural isolation barrier might be sufficient.

It is the author's assertion that, for this Model Regime, if the Host requires sanitization of equipment, then C&S system designs should allow for *in situ* verification of the physical status of the item under inspection, with further data collected through an isolating barrier or sacrificial component. This relates more to inspection of equipment, rather than inspection of a TAI.

Significantly, the retrieval complication also has implications for transferring C&S data back to Inspectors for analysis. If the medium of data transfer itself is not isolated from the environment, it too will be considered for sanitization, and so C&S system designs should also consider how to isolate data media, or otherwise protect inspection data

Facility Health and Safety

Any environment in which explosives are present will require limitations on the risks posed by use of electronics or other equipment that could deliver energy into sensitive circuits or explosive materials. The use of technologies that broadcast signals could be restricted. Host Certification requirements extend to ensure all technologies deployed into facilities are safe within that environment, and so equipment designs must also factor this in as a necessity. Using electronic tags such as Radio Frequency Identification (RFID) Tags is problematic because of this.

Safety and security considerations also affect the time windows and time limits available for Inspection activities. Dismantlement operations cannot be delayed or postponed with TAIs in exposed configurations. The time available to review containment and surveillance data, draw conclusions and act upon them will therefore be limited.

Authentication in the Model Regime

It is reasonable to assume that, comparably with Safeguards, the Inspecting Party must be able to establish and maintain trust in all equipment used, and data generated, in support of the verification regime, even when equipment is left within the Host facility for some considerable amount of time. Authentication is a general term for the tests, processes and procedures that must be developed to foster the necessary trust in Inspection equipment.

Authentication is required to ensure that equipment operates as designed and remains in a pristine state, so that, operating in its agreed capacity; the data collected 'accurately reflects the true state of the monitored item' [7,8]. This is a complex process that could be more difficult to achieve in the model VWDR Regime, given Host Certification requirements and control over access to Certified equipment.

Direct Authentication

Authentication measures applied directly to the unit of equipment that is to be deployed are here referred to as 'Direct Authentication', and could be restricted. Direct Authentication could lead to a permanent cycle of Certification and Authentication as both Parties attempt to establish trust in

equipment once left in the other's possession, so a method of terminating that cycle is required. Since, in this Model, the Host will simply not allow equipment into a sensitive facility until certified, it is Authentication processes that must be adapted.

Containment and Surveillance systems designs should consider that:

- The Inspecting Party may not be given sole access to equipment that will enter the facility.
- Any tools and equipment required to authenticate inspection equipment will also have to be certified by the Host for use with the Inspection equipment, if used on equipment that will enter the facility.
- Direct Authentication will have to take place under secure conditions.

In light of this, Direct Authentication measures might well be restricted to techniques such as functional testing; with further tests possible utilising Host provided tools.

Authentication by Association

With the scope of Direct Authentication restricted, 'Authentication by Association' may be the most common approach for the Inspectorate to gain confidence in the equipment they are using. The approach is often described as 'random selection' [9] and would involve the Host Party providing many 'identical' copies of certified equipment, some of which are then chosen at random by the Inspecting party to go into the facility, and some of which are chosen to be taken away and authenticated against the agreed standard design, in any way deemed necessary. Those units chosen to go into the facility must remain under joint custody at all times, to ensure both Parties are confident that they remain in pristine condition.

Authentication by Association is obviously not as robust as Direct Authentication. There is therefore a second driver to develop robust simple technologies that can be tested by the inspecting party to a satisfactory standard, even under the strict environment resulting from the conditions of this Model Regime. 'Simple' in this respect describes technologies with capabilities and functions limited to only those absolutely necessary to perform the primary, intended purpose of that technology. Authentication by Association may then still provide a supporting function.

Post-Use Authentication

If it is possible to demonstrably isolate containment and surveillance equipment from the facility during Certification, it might be possible to retrieve that equipment from the facility post-deployment. Use of environmental isolation 'barriers' or 'jackets' could enable the actual deployed unit to be forensically analysed for inspection, verification and Direct Authentication purposes post deployment by the Inspectorate, as opposed to relying on Authentication by Association. Once retrieved, the unit could not be deployed again within the facility, unless first recertified.

Electronic equipment, designed for post-use Authentication will have to account for the possibility of the manufacturer adding false data and self deleting routines into the processing unit in such a way that false data replaced genuine data, though functional testing and suitable calibration techniques could mitigate this threat to some degree.

Post use Authentication techniques could prove a useful option for some systems, though some technologies will not lend themselves to being suitably isolated from the facility as to do so would affect the data being collected.

Dynamic Disassembly Operations

During warhead dismantlement, the configuration of material will change over relatively short timescales. Mass balance (if allowed) and NDA measurements cannot effectively account for *all* the material going through a disassembly process, or once the configuration of the TAI has changed. If the inspection data collected warrants further questioning, notification of this will be of most use whilst the TAI can still be directly associated with the stage of the dismantlement process to which the discrepancy relates.

In order to maintain the chain of custody over all material contained within process area boundaries, containment and surveillance data needs to be collected quickly and analysed sufficiently within the dismantlement process timeframe, so C&S systems must be configured to enable rapid analysis.

Though there is a burden upon the Inspectorate to ensure data analysis occurs relatively quickly, there is might also be an advantage not present in other regimes, at least during the dynamic, dismantlement stage of a VEDR: Equipment could be deployed and left unattended for shorter timescales than in other situations, leaving it vulnerable to attack for less time. This may affect, for instance, the level of complexity required in a unique tag to assure its own authenticity.

Summary of Model Effects

A summary of some of the operational constraints effecting implementation of a Chain of Custody within the Model Warhead Disassembly Verification Regime follows:

- The Inspecting party agree with the Host facility the *design* of equipment that can be deployed within that facility. All equipment must adhere strictly to jointly agreed standards, with exact specifications for all components. Any change to that design may require Certification and Authentication.

- Strict, joint design standards mean use of either covert or complex features do not provide a method of assuring the Inspectorate that equipment is genuine, or that it functions as expected.

- The Host will not allow the Inspecting Party to provide equipment for its own use within sensitive facilities, without it undergoing an indefinite period of testing for Certification. Testing may be destructive, or require disassembly and rebuilding of the item. Full disclosure of the design would be necessary. Rather than face the uncertainty of this testing process, it is likely the Host will manufacture or otherwise provide all equipment used in such facilities.

- A system that can be segregated into components that either require or do not require access to sensitive facilities may be beneficial. Data diodes should be incorporated into such systems.

- The Inspecting party cannot trust data emanating from equipment until the equipment has been authenticated, but Direct Authentication techniques will be limited on units of equipment entering sensitive facilities. Authentication by Association with ‘identical units’ may feature heavily, or post-use Authentication *if* the system has been designed for this purpose.

- Applying complex Authentication techniques may have to take place under secure conditions, if performed on equipment destined for use within the facility. Certified equipment must be recertified if left solely in Inspector custody.

- The Inspectorate may not be able to simply retrieve items from a sensitive facility. Methods of data collection and retrieval, and equipment inspection procedures, must take this into account. The system design may have to incorporate sacrificial components; because facility will privately sanitize any item if it may have collected sensitive information from within the facility. Physical isolation from the surrounding environment may be necessary if that item is later required outside of the facility. Data transferral methods must consider this.

- Classified data or information relating to the weapons systems themselves, facility operations or the physical security of the facility cannot leave the site, unless explicitly agreed by the Host. Interrogation techniques that might collect such’ peripheral’ data will be curtailed. System design and operation should ensure no such data could be collected.

- Data may be reviewed by the facility prior to it being released to the Inspectorate, if the equipment used could have recorded security-sensitive information. If classified details are recorded, the data will not be released to the Inspectorate.

- Technologies may be further restricted to minimise external sources of energy, particularly around explosive materials. Power sources should be isolated from facilities and intrinsically safe. Technologies that generate electromagnetic fields may be a concern. RFID is not a suitable solution for multiple reasons.

- The Inspecting Party will have a limited amount of time to collect and analyse containment and surveillance data, if they are to maintain custody over all the material resulting from dismantlement.

Tagging System Design Considerations under a VWDR

The utility and security of existing systems should be re-evaluated under the different operational parameters of a restrictive Verified Warhead Dismantlement Regime, as summarised in the above Model. A brief discussion of a high security tagging technique follows, in order to explore operational concepts and design considerations for any system.

Tagging Systems Based Upon Random Distributions of Particulate

The Reflective Particle Tag, developed by Sandia National Laboratories, is a secure tagging technology. The concept is versatile, could be incorporated into many different applications, and meets many of the requirements established in the Model, for use during a VWDR.

Reflective Particle Tagging requires a reference image to be recorded, of a random distribution of particulate encapsulated within a stable medium. A second image is then recorded at a later time, and a comparison method is used to determine whether the two images match. The high security element arises from the reflective and other physical characteristics of the particulate used, combined with controlled conditions during photography of the tag.

RPT based systems may require operational protocols that differ according to the exact system configuration, but multiple configurations could maximize its utility. Discussion of such a protocol demonstrates well the equipment configuration and operational choices faced if tasked with implementing any tagging system.

Generating the Tag

Drawing from the Model, a Tagging system suitable for a VWDR should account for the fact that the Host may effectively supply all inspection equipment, and for the fact that the Inspector may have limited opportunity to authenticate the item deployed into the facility. The location at which a Tag is created may have suitably controlled by both Parties, so ensuring a tag itself is unique is therefore best performed by the Inspectors generating that tag from certified raw materials supplied by the Host, once inside the controlled environment of the secure facility.

Creating random particulate tags requires a suitable matrix and suitable particulate. A mutually agreed design could specify those materials, whilst accounting for facility regulations and vulnerability concerns. Either party might actually supply the raw materials. Authentication or Certification of a random sample from a batch of the materials could ensure the entire batch is compliant with the joint design. The Inspecting Party could then generate tags prior to deployment, guaranteeing that the RPT tags are unique.

The creation of a unique tag that can be trusted by all Parties can be achieved relatively simply.

Data Capture

A camera is needed in order to image the random distribution of particulate in the tag. This discussion presumes a digital camera of some type would be preferable in order to compare large amounts of data quickly.

The Model indicates that it is beneficial to separate system components, so that the minimum numbers of devices need certification to enter sensitive facilities. A camera for tag data capture can be

separated from the data analysis equipment. This might offer the benefit to Inspectors of more flexibility concerning the provision, Certification and Authentication of the analysis equipment, so long as it need not enter sensitive facilities within the disassembly complex. For this system configuration to be certifiable, the system must incorporate a data diode.

In order to authenticate a selected camera, functional testing could go some way to ensuring it operated in the expected manner, and that the data received from it was a true image of the subject photographed. This could be combined with more rigorous Authentication by Association, requiring the availability of multiple cameras for random selection prior to deployment. A modular data capture system which allows the randomly chosen camera to be incorporated at any time is therefore preferable in this case. The camera would effectively become a sacrificial component.

A removable isolating barrier or jacket could encompass an entire camera system, in much the same way as commercial waterproof cases are designed for underwater photography. The entire camera system could then be subject to post-use Authentication measures, once that camera was no longer required. In this iteration of a design, the modular components may consist of the new data storage devices and a new isolation barriers required in order to use the camera each time it entered sensitive facilities.

Cameras are versatile and complex instruments, so the Host will not consent to Inspectors freely using them within the facility. Data collected by the camera also cannot be directly released to the Inspectorate if there is a possibility that sensitive data has been recorded, but a combination of system design and operational procedures can be taken to address this.

In accordance with the Model Regime, all aspects of the standard camera design need to be jointly agreed, to enable Certification and Authentication. The Host may insist on manufacturing or sourcing camera systems that meet the agreed design, guaranteeing the absence of hidden sensors unconnected with the primary function of the camera. Operation of the camera could also be restricted to facility personnel, to reduce the possibility of the camera being used for other purposes. However, this situation may also benefit the Inspectorate because data arising from that camera could then be considered for release directly back to Inspectors, so long as it is transferred in a suitable fashion. The combination of Host provision and operation might provide the necessary assurance that the data is suitable for release.

If it is necessary for the host to operate equipment in order for data to be released in a satisfactory manner, the design of the tagging system must take this into account. If analysis of the tag requires data to be captured in a specific, controlled manner, the system design should ensure repeatability of data collection no matter who operates the camera.

Commercially available cameras have many functions that are peripheral to the actual function the camera is required to perform in this case; so all additional functionality should be disabled or designed out, if use of commercial cameras is preferred. Limiting capabilities will make the camera easier to certify and authenticate. If a commercial camera is used, full disclosure of the design details may be necessary to establish the agreed ‘joint’ design.

Additionally, the camera system will have to meet all safety requirements stipulated by the Host. If used around explosives, aspects such as power supply, isolation, and output will need to be jointly addressed.

Design of data capture technologies will require intense of cooperation between the Host Party and the Inspectorate if they are to be Certifiable and Authenticable. Operational procedures should be discussed early in the design, to ensure the system will deliver quality data in a restrictive environment. Full disclosure of designs might limit use of commercially available hardware, which may need significant modification to reduce superfluous functions that might otherwise be manipulated to gather data.

Data Retrieval

Methods available for extracting the data from the camera could be affected by facility sanitisation requirements. The options for transferring data might include:

- A wireless or wired data link, allowing data to be downloaded 'live' without physical contact between the camera data or memory device, and personnel from either Party
- Physical removal of an data storage device from the camera

'Live' Data Downloading

Data downloading options could potentially include an IR link or 'Bluetooth', or a data cable.

A tagging system that incorporates any such method would require a data receiver, but could be designed so that it need never enter sensitive facilities. Environmental concerns could be addressed by positioning the receiver behind an isolation barrier on the edge of sensitive facilities, so there may be no need for sanitization of the data receiver. The camera itself would in effect become a sacrificial component of the tagging system, but the benefit may arise from data that is released without physical handling by the Host.

To enable direct downloading of data, the data receiver would have to be certified and authenticated to the same level as the camera itself and may also have to be designed in such a way as to demonstrate a data diode, to ensure information could only flow from the camera to the recording device.

The method of transfer must also be compatible with facility explosives regulations, so the system design may incorporate power restrictions, along with component configurations and operating instructions which require the 'camera end' to be transferred to a designated zone for safe data transferral.

Sending the data wirelessly or through a cable could introduce vulnerabilities that may require each photograph to be digitally signed by the Inspectorate. If the data requires a digital signature, suitably certified equipment must then also be introduced to enable this. The Host may have had total control over the hardware required to generate the Inspector's secure digital signature.

This burden of Certification and Authentication requirements is huge, complex and extends across all equipment that links to the camera itself or enters sensitive facilities. A system based upon data downloading would require suitably certified and authenticated cameras, data receivers, and encryption hardware necessary for digitally signing the data. A data download link therefore has challenges that require much consideration. The Certification and Authentication requirements may make this type of system unsuitable.

It may be that, in order for generation of encryption keys to be performed by the inspectorate, the system would have to be designed in such a way as to demonstrate to the satisfaction of the Host that no matter what information was actually contained on the encryption key, no alteration in the function of the inspection hardware could occur. The hardware design might have to demonstrate that reprogramming was impossible. The Host may also still insist on providing the media hardware used for transporting and connecting the key to the hardware containing the data to be signed.

The second option is physical removal of a data storage device housed within the camera.

Physical Removal of Storage Media

By physically removing the data storage device from the collection equipment, the quantity of items requiring Certification and Authentication could be reduced. The physical removal of the memory device also acts as the data diode; once the memory device had been handed over to the Inspectorate, it would not return again for use with certified equipment.

In order for a memory device to be removed and handed directly to Inspectors, the design of the camera system must include measures to ensure the device remains physically isolated from the facility environment at all times. A system incorporating an 'isolation jacket' to enclose the camera and the memory device is therefore attractive from this perspective also. Removal of the isolation measures employed would need to take place away from sensitive facilities. This might allow for the data storage device to be transferred into Inspector custody with no further Host control over it. Using this technique, the Isolation barrier becomes the sacrificial component.

It the Data Storage Device isn't isolated from the environment the data might still require a digital signature, to provide protection during sanitization of the memory device itself. An alternative might be to copy data from a 'dirty' data device to an isolated device, but this may require a further piece of complex, host provided equipment that would need Authenticating.

Data Analysis

The Authentication requirements placed upon the Inspectorate may be too great if the analysis hardware and software were to be provided by the Host. Separation of camera and analysis system is therefore very important, and can be accomplished for a tagging system using particulate distributions. Data analysis, even in the Model conditions, could possibly therefore take place away from sensitive facilities. So long as the Host is happy to release the data, the Author asserts that analysis could possibly take place on Inspector provided equipment, in this case.

Many identity analysis techniques are automated, and are designed to measure the similarity between reference and verification data. This allows large volumes of data to be processed quickly, which suits the requirement to analyse data collected within a relatively narrow timeframe.

Automatic analysis systems may be configured to minimize false positives and false negatives, i.e. to minimize instances where different tags are accepted as the same tag, or instances where data concerning the same tag is not verified; but an automated system will treat well counterfeited or subtlety changed tags with the benefit of the doubt because of the tolerances necessary for designating matches between images.

Whilst tag data analysis systems look for similarity between reference and verification data, there is therefore an opportunity to counterfeit the tag. With complete knowledge of the tagging system, the Host will know the threshold a counterfeit must pass, if it is to be automatically accepted.

Having an Inspector undertake the analysis themselves would slow the process down and would inevitably become more subjective, but would enable a much more subtle analysis, better able to pick up on signs of counterfeiting and possibly to account for differences due to misalignment between camera and tag.

To be most useful, automatic analysis techniques may benefit from highlighting differences in tags, even if they are sufficiently similar to pass analysis. The option is then available for the Inspector to become involved in analysing tags when necessary, but the overall speed of the system need not be compromised to the extent a fully manual system would require.

For this tagging system, alignment tolerances can be reduced if the design includes a method of physically attaching the camera to the tag, to ensure the position of the camera in relation to the tag is as similar as possible between images. The method of attachment and alignment will obviously have to consider how and where the tag is to be deployed.

The threat of data manipulation extends when the Host controls equipment; all data could be manipulated at the collection point in such a way as to obscure skilled attacks or deliver useless data. It is therefore even more important to ensure data collection methods are standardised to ensure repeatability in data collection, if the analysis technique requires precise measurements.

Summary of Tagging Considerations

The discussion demonstrates how quickly consideration of a specific technology requires a solution to general concepts. Even a notionally simple tagging technique requires much thought about the implementation of the system. Generating the tag is the simple part. Retrieving the data in a useful and mutually acceptable way requires a lot of attention to operational procedures, the extent of interacting technologies and the burden each technology adds to the Certification and Authentication measures required.

The tagging system must provide trusted data, even if the Host provides all materials and equipment for deployment into sensitive facilities, and data collection method must be repeatable, no matter who operates the system.

A ‘simple’ tagging system based on random distributions of particulate may be able to meet the stringent requirements of a VWDR in a number of ways, but any solution will have to be tailored to the specific Regime requirements. Each design choice will affect further design choices and the operational procedures of each component.

The tag itself can be generated relatively easily but the versatility of the tag may be reduced by the need to align the camera. Data retrieval could be accomplished in a variety of ways, some of which may not need data to be digitally signed but would require strict operational protocols. For those that do need signing, more work is required to develop satisfactory methods of Certifying and Authenticating the technologies and processes required to achieve this. The tagging system may require some component to be sacrificed to the facility, so it may be better to incorporate a disposable jacket into the design for this purpose, as a cheap, non-critical solution.

The method of analysing the data should be designed to alert the Inspectorate if a subtle deviation from the expected value is found, to allow for more detailed analysis.

Conclusion

The paper has explored some of the reasons why a Verified Warhead Dismantlement Regime could be considered a new operational paradigm. The requirement for the Host facility to first Certify all equipment deployed into sensitive facilities has ramifications throughout the whole design process for inspection equipment, including containment and surveillance systems.

Even ‘simple’ acts such as tagging an object require a detailed analysis of how the restrictive conditions of a VWDR might affect the suitability of existing methods of inspecting and analysing that tag. Similar thought processes could be applied to many systems that might be required in such a regime. Existing equipment, operations and concepts cannot necessarily be transferred across from other regimes; to do so may open them up to vulnerabilities not present elsewhere. The Host may not allow some techniques to be utilised within security-sensitive facilities. Existing technologies may not be certified without full disclosure of design details, and possibly without restrictions upon current capabilities.

Though the Authentication and Certification requirements are huge for a Verified Warhead Dismantlement Regime, there is the possibility they could be influenced by operational procedures. System Designs should consider how to separate the total system into component parts; how to minimise superfluous functions; and how to ensure data can only flow in one direction, in order to further reduce the Certification and Authentication burden. At present, Certification and Authentication techniques represent significant technical challenges that are still to be addressed, even for ‘simple’ technologies such as passive tags.

Appendix I

Limits to the use of Non Destructive Assay and Information Barriers

Non Destructive Assay can be defined as any measurement or interrogation technique used to obtain data about an item without recourse to removing a sample of material from it, and, when applied to nuclear material, usually implies measurement of radiological emanations arising from nuclear processes occurring within the material.

NDA of the Treaty Accountable item could play an important role in verifying aspects of a warhead undergoing dismantlement to ensure the item presented meets exhibits the agreed characteristics of a warhead, but there are limits to its effectiveness within a warhead dismantlement verification regime for fissile material accountancy purposes.

The limits arise because of the need to protect classified information, which in turn limits the amount of information declared about any given warhead system. Typically the declaration could expect to include:

- That it contains a certain material
- That the isotopic ratio of that material is consistent with a weapon system
- A value for a minimum mass of material, indicative of material presence, but insufficient to define actual quantities present. (The declared value could be significantly lower than those values defined as 'significant quantities' under Safeguards regimes).

With such vague limits set and the level of detail inherent in NDA data, all NDA data analysis must take place behind an information barrier. Information Barriers have been much discussed elsewhere [10, 11]. For the purposes of this paper, suffice it to say that NDA results will be important but of limited value if performed in the absence of a robust chain of custody over the item being dismantled.

An attribute measurement system will only confirm that the TAI meets the minimum threshold of declared attributes, though a good IB should be able to discount the presence of spoofing materials. It will not be able to tell the Inspecting Party whether *all* of the Treaty Accountable Material initially present in the warhead is still present after dismantlement.

As the TAI continues through the dismantlement process the configuration (& therefore NDA signatures) of the TAI may change. A template NDA measurement at the beginning of a dismantlement process will therefore also fail to provide confidence that *all* of the material is still present after a dismantlement process.

1. A Treaty Accountable Item could be defined as any item declared by the Host as meeting the characteristics used to identify an object as nuclear weapon, as agreed by the Parties within the Regime Treaty. Typically a TAI could be a complete warhead, a partially dismantled warhead, or the fissile material arising from dismantlement.

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3. Merkle, Peter B., Horak, Karl E. e*t al*, '*Automated Reflective Particle Tag System for Physical Authentication*', Proceedings of the INMM Annual Conference, Baltimore, MD, USA, July 2010

4. Supply in this instance means either actual manufacture of the equipment or the unlimited possession and analysis of any component of every unit of equipment supplied by the Inspecting Party. Given that the Host has all standardised equipment designs, identical replacements can always be introduced.

5. A Data Diode would be a physical or procedural method of ensuring data can only flow from certified equipment to a designated receiver, ensuring the certified equipment cannot receive modified operating instructions from any outside source.

6. Johnston, Roger G., and Warner, Jon S., '*Unconventional Approaches to Chain of Custody and Verification*', Proceedings of the INMM Annual Conference, Baltimore, MD, USA, July 2010

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Introduction to Attributes Information Barrier for Nuclear Warhead Authentication

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Abstract:

Part of a chain-of-custody approach to warhead dismantlement verification is the authentication process, where nuclear measurements such as gamma spectroscopy and neutron counting are undertaken to confirm the identity of a warhead. Direct measurements could reveal sensitive data. Information barriers can be used to deliver an unclassified output of classified data measurements. This paper introduces a project which develops an information barrier using an attribute approach.

It analyzes what information could be revealed during direct measurements, explores different information barrier concepts and information barriers built in the past to prevent leakage of sensitive information, suggests a warhead authentication procedure, introduces attributes that could be defined to authenticate a weapon and suggests measurement techniques for each. This paper is an introduction of the project; results are preliminary as the project will be carried out in the future.

Keywords: disarmament verification**;** information barrier; attributes; warhead authentication

1. Introduction

One measure that builds confidence in nuclear dismantlement is the so-called chain-of-custody approach. This chain-of-custody "demonstrates that an unaltered or uninterrupted custody or control of an item has been maintained by the owner or inspector, [...] that provides confidence that deceptions have not been introduced" [1]. This means that a verification regime must provide confidence that from the very beginning of the dismantlement process of a nuclear weapon to the very end no cheating has taken place. While some verification measures have been implemented in previous treaties, a verification system that enables a complete chain-of-custody approach is not available today. A very important part is the actual dismantlement of the warhead - the moment where the warhead itself is taken apart. In order to guarantee a complete chain-of-custody, it must be assured that the device that is being dismantled is in fact a nuclear warhead, the so-called warhead authentication: "Successful authentication will ensure the monitor that accurate and reliable information is provided by a measurement system and that irregularities, including hidden features, are detected" [2]. During the warhead authentication process, it will be assessed whether the warhead is a true warhead or rather a dummy. Such a system is not used today and technology development has been limited so far.

A warhead authentication system can more readily be agreed on if the measurement system does not reveal sensitive information on specific warhead design features as nuclear weapon states consider

this information to be highly classified. Therefore, an information barrier (IB) could be used that would analyze measurement results in an automated manner and produce an unclassified output.

The project introduced here tries to develop a warhead authentication system using such an information barrier. This paper is merely an introduction into the topic as the project is still in progress – it presents a number of concepts and measurement tools that could be useful.

2. Revealing sensitive information during dismantlement verification

It must be identified what information could possibly be leaked during the verification process. The verification system developed in this research uses gamma spectroscopy and neutron counting. However, the gamma spectrum will give experienced people, for example (former) weapon designers, the possibility of reverse-engineering - at least to some extent. Most obviously, the isotopic composition of the fissile material can be deduced. From this, it can be determined if plutonium, uranium or a mix of both is used in the fissile core. Also, information regarding enrichment, age (e.g. date of plutonium separation) and further properties of the material (e.g. presence of other isotopes such as U-232 which would indicate that the uranium was used in reactors before) can be deduced from the isotopic composition. This list is by no means exhausting. Neutron counting might give more quantitative information. Under special circumstances where some information regarding the warhead design is initially available to inspectors, a rough assessment of the fissile material mass might be possible. However, information regarding shielding of radiation emitted from the core (see below) is required as well as the isotopic composition.

Apart from information regarding the fissile core, further design features could be reverse-engineered. By conducting a number of measurements in different positions relative to the warhead, information regarding the form of the warhead (e.g. degree of spherical symmetry) could be revealed. Also, information about other materials present such as a neutron reflector or a tamper could be exposed. Knowing the neutron cross-sections and gamma attenuation coefficients of absorbing materials which vary at different energies, a well calibrated detector could reveal this information, when peaks at different energies (in the case of gamma spectroscopy) are analyzed with regard to the measured and expected intensities in relation to each other. An assessment of the thickness of the absorbing material can also be given once the absorbing material has been identified by comparing peaks at different energies, see for example [3].

It is important to note that reverse-engineering requires a lot of experience. Especially in the reverse-engineering of absorbing material, expertise is needed that inspectors might not have. It is therefore not easy to assess how much sensitive information could be revealed.

3. Information barrier (IB) setup

3.1. Requirements

The IB is a computing system that receives the raw classified data and calculates the unclassified output. The system has to protect the information from unauthorized access by the inspector. Also, the data collection and processing equipment must be protected from the inspected party so that no tampering with the measurement system is possible. This means that input possibilities of both the inspected and inspecting party should be reduced to an absolute required minimum.

After initial authentication (testing), the system should be designed in a way to minimize the possibility of alteration and maximize the detectability of alteration by any party. The system should have a simple design that is easy to check and understand.

As a last major requirement, as few sensitive data should be stored as possible for the shortest time possible. No permanent storage of sensitive data should be necessary because in case the system is tampered with, sensitive data could possibly be extracted. This danger should be minimized which especially sets the requirement that no sensitive data should be stored which could be assessed even in between inspections.

3.2. Solutions

The Trilateral (US-Russian-IAEA) Initiative that was conducted between 1996 and 2002 aimed to establish a system of verification under which states in possession of nuclear weapons might submit excess fissile material to IAEA monitoring [1]. During the Trilateral Initiative, the so-called “Attribute Verification System with an Information Barrier Utilizing Neutron Multiplicity Counting and High-Resolution gamma ray Spectrometry” (AVNG) has been developed. This system is designed to contain a minimal non-volatile memory; all classified data are stored on volatile memory. This means that after power shut-down which could either happen during inspection if irregularities are discovered or after the inspection has been performed, all sensitive data will automatically be deleted. The software resides on non-rewritable programmable read-only memories [4]. This is to ensure that no alteration of the algorithm is possible and there is also no possibility to obtain access to classified information. Furthermore, there are neither hard drives, nor other mechanical drives, nor network capabilities in the computers. The only possible operator input is through simple switches. All measurements are controlled by these switches and to further minimize possibilities to interact with the system, it is specified to include appropriate shields and other devices to prevent transmission of electromagnetic signals into (to alter it) or out (to reveal classified information) of the system [4]. The AVNG can operate in an open and in a secure mode with the latter one being the default. In the open mode, access is given to all raw data while in the secure mode - designed for the verification of classified materials - only unclassified outputs will be given. The open mode could be used to search for errors and to solve them. A “security watchdog” overlooks the modes of operation. This device controls the system in combination with an emergency “scram” switch [4]. This system would shut-off power when an intrusion into the system is detected. This results in complete information loss in the volatile memory. The system's security functions are separated from the measurement function. There is no direct communication possible between security watchdog and measurement/analysis system except for power [4]. Therefore, measurement equipment cannot be influenced.

A physical IB was also built during the Fissile Material Transparency Technology Demonstration (FMTTD) performed at Los Alamos National Laboratory. It has a very similar design as the AVNG. It has a shielded electronics rack with a sensor at the door. When doors are closed, only a green or red LED is visible to indicate binary outputs. Once the door is opened, all power is automatically removed from the system [5]. A scheme of the entire system is provided in Fig. 1.

The detector systems transmit the classified measurement data to the computational block that applies the algorithm to obtain the unclassified output. The next step is the data barrier (which is not the same as the IB introduced in this paper). Its function is to pass the unclassified information in only one direction (towards the LED output) and prevents information flow in the other direction [5]. It also disallows passage of classified information. This is a double check since under normal circumstances, only unclassified information passes the data barrier anyway.

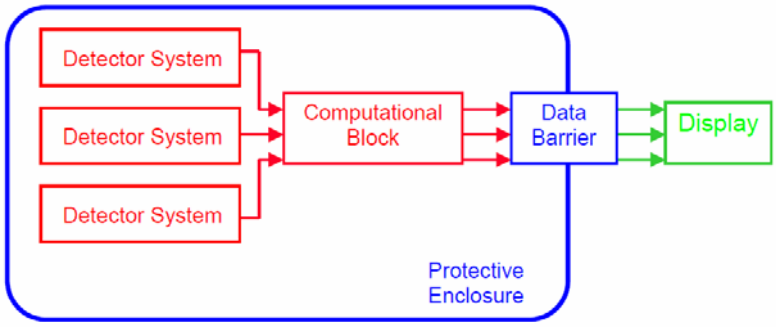

Figure 1: Attribute Measurement System with Information Barrier scheme that shows the attribute measurement detectors, the attribute-analysis computational block and the data barrier [5].

4. Information barrier concept

The IB prevents the leakage of classified information but at the same time gives confidence the system is performing: Detailed and sensitive measurements will be performed, but the result will be delivered in unclassified output (for example a binary “yes”/”no” output). There are two basic concepts that will be described in this section, the template and the attribute approach.

4.1. Templates

In the template approach, properties of all existing warhead designs will be measured and stored. For example, this could be a gamma ray spectrum of the warhead containing detailed and sensitive information. The inspecting party should never be able to have access to the stored data; in addition, it has to be guaranteed that there is no possibility for the inspected party to alter the stored data. Before dismantling a warhead of a certain design, a gamma spectrum will be recorded and the IB will compare the recorded spectrum with the stored data. If the two spectra match within a certain deviation, the IB output will display a “yes” signal, if there is no match it will display “no”. This procedure does not need to be limited to gamma spectroscopy, but the approach can possibly be applied to other measurements as well. It If a template IB works correctly than the deviation of a measurement compared to the template is small enough to ensure that dummies will be detected, but large enough to ensure that real warheads will not be detected as dummies (false negative).

The U.S. Department of Energy has sponsored the development of template IBs and tested their reliabilities. Brookhaven National Laboratory developed the Controlled Intrusiveness Verification Technology (CIVET) which was modified by Sandia National Laboratories for use with the Radiation Identification System to produce the Trusted Radiation Identification System (TRIS). Tests of TRIS conducted at the Pantex plant confirmed that it is able to identify various types of weapons and weapon components. During the test, templates for five weapon types were able to identify the corresponding five weapon types during the test that were previously unknown to the inspectors [6].

The main advantage of the template approach is that it is considered impossible to design a dummy warhead that matches within uncertainties with a particular warhead in the template since it is very difficult to reproduce the identical comprehensive characteristic spectrum. Since the exact signatures of the recorded warhead are given, only small deviations will be detectable. The probability of dummy warheads not being detected is very small.

There are also significant disadvantages with a template information barrier. First but foremost, the inspecting party has to have confidence that the warhead to serve as a template is indeed a warhead. The inspecting party needs confidence that the inspected party does not cheat during this initial step. One option would be to require measurements from a number of weapons with the same design that are randomly chosen by the inspecting party [7]. A separate information barrier could analyse the templates in regard to their deviation from each other. If it turns out that one of the weapons has a significantly different gamma spectrum for example, it might be considered that this was not a true warhead. Measuring a larger number of warheads would make a cheating scenario easier to be discovered. The final template to be used could be the average of the measured warheads; in addition measuring a number of warheads and combining them into a template already gives a measure of standard deviation of single warheads from the template. But even when randomly choosing a number of warheads, there is still no full guarantee that the devices were actually warheads. Another very significant disadvantage is the storage of the template. The template which contains very sensitive information on the specific warhead design has to be permanently stored on a storage medium. It must be ensured that there is no possibility for the inspecting or even third parties to gain access to it. Furthermore, there must not be a possibility for the inspected party to make changes to the data. This could be done by placing the storage medium in a safe that requires two combinations - one in possession of the inspecting and one in possession of the inspected party - to open it. Additional protection could be provided by encrypting the data with a two-part cryptographic key (again one for the inspected and one for the inspecting party). An alternative is to give the storage medium to the inspected party while the IB would provide the inspecting party with a digest or secure hash of the template when it is made and each time it is used. It would unambiguously confirm the template's authenticity [8]. If a number of warheads were measured for the template, an assessment of the variations exist. If, however, only one warhead was used for the template, it is not known how much other warheads of the same type deviate. Such an estimate is required because it must be defined

what level of deviation from the template is allowed and what level indicates a dummy. This could be hard to define.

4.2. Attributes

In the attribute approach, no reference data is needed. Instead, the two parties agree on a set of attributes that define a nuclear warhead. All nuclear warheads will in an ideal situation fulfil the attributes, while no object other than a nuclear warhead will. Attributes can relate to nuclear measurements (for example gamma spectroscopy and neutron counting) and to non-nuclear measurements as well. There is the possibility of defining quantitative values for each attribute and acceptable deviations; another possibility is defining an allowable range of values, for example minimum or maximum allowable thresholds [9]. Crucial to an attribute IB is the development and agreement among the parties involved on the algorithm of the IB. Again, the output of the IB could be a “yes” or “no”. Different output options (such as a deviation from threshold or mean values or a fuzzy logic approach that yields an index) might be considered as long as they prevent sensitive information from being revealed. On the one hand, a binary output is the best solution from the point of view of not exposing sensitive information, while on the other hand, different outputs might be preferred from a point of view of giving the inspector sufficient information to reach a conclusion regarding the authenticity of the presented warhead.

The Trilateral Initiative developed an attribute IB. Although detailed descriptions are not publicly available, some technical information was released [4]. In 2000, the FMTTD was performed at Los Alamos National Laboratory, where the determination of attributes of unclassified plutonium and a US nuclear weapon component was demonstrated . The IB was sufficiently robust to allow measurement of a classified weapon component without revealing classified information[1]. Some technical information in regard to this project can be found in [5, 10, 11]. During the U.K.-Norway Initiative, IB designs were also developed [12]. This initiative was a project between the U.K. and Norway, where a warhead dismantlement verification simulation exercise was performed, see [1]. However, instead of a nuclear weapon or a material that could somehow be significant for nuclear weapons dismantlement verification, a Co-60 source was analyzed which only has limited technical relevance when it comes to nuclear weapons dismantlement verification.

In comparison to template IBs, the most significant advantage is that no data has to be stored permanently. Measurement results can be stored on volatile memory which is erased after power shut-down. This significantly decreases the danger that classified information will be released.

Attributes IB also have a number of disadvantages: Depending on the quality of attribute definitions, there might be the possibility that a dummy warhead could result in an IB output indicating that it is an actual warhead. It is a challenge to create and agree on a set of attributes where cheating becomes virtually impossible. That said, it will still be a tremendous challenge for an inspected party to cheat - especially in the case of a very comprehensive set of attributes.

This project will deal with the attribute approach and not with the template approach because of very limited knowledge about nuclear weapons design that is publicly available. For independent research on the template approach, detailed design information is necessary which does not exist in public. This is not the case for the attribute approach where more general (and available) knowledge is sufficient.

5. Effect of shielding

Performing necessary technical steps to authenticate a nuclear weapon at the nuclear weapon itself prior to its dismantlement poses significant technical difficulties. These difficulties mainly come from the shielding of the nuclear weapon pit containing the fissile material. Naturally, most attributes would be related to verifying certain characteristics of the fissile material in the weapon. Because exact nuclear weapon design details are neither available for independent researchers, nor for inspectors

[1] see http://www.lanl.gov/orgs/n/n1/FMTTD/index_main.htm for more information on the project including technical papers with descriptions of the used components as well as lessons learned.

performing verification measures, it is fairly unknown what surrounds the fissile material. It is unknown how much material (and which) acts as shielding of radiation. This information is however important for assessing quantitative information.

This paper does not intend to focus on the design of nuclear weapons. While little information has been de-classified, a number of papers exist in open literature. The fissile material of a nuclear weapon could possibly be surrounded by a tamper, which delays the expansion of the fissile material after the fissile mass becomes critical. A neutron reflector could be used to increase the yield. Depleted uranium or tungsten / tungsten carbide could be used among others; neutron reflectors could be made of beryllium. Nuclear weapons need conventional high explosives to initiate nuclear fission that contain large amounts of hydrogen, carbon, oxygen and nitrogen [13]. Advanced weapons make use of a primary (similar to a fission weapon described above), but also include a secondary (fusion stage).

Gamma rays are effectively attenuated by high-Z materials (Z being the atomic number) which are very likely to be present in some warheads (e.g. uranium or tungsten):

$$I = I_0 \cdot e^{-\mu \rho L}$$

where I_0 is the original gamma radiation intensity, I is the intensity after attenuation by attenuation coefficient μ over length L and absorbing material density ρ [14]. The photoelectric mass attenuation coefficient τ is given as [14]

$$\tau \propto \frac{Z^4}{E^3}$$

Neutrons can also be shielded. Neutrons can react via elastic scattering, neutron-induced nuclear reactions or inelastic scattering. Elastic scattering slows them down and changes their direction [15]. Neutron-induced reactions include the radiative capture reaction (n,γ). The probability of many neutron-induced reactions drops off rapidly with increasing neutron energy. In elastic scattering, the neutron can transfer an appreciable amount of energy in one collision. The average energy loss due to elastic scattering is given as $2E \cdot A/(A+1)^2$, where A is the mass number of the nucleus and E the energy of the neutron [14]. It is clear that the energy loss decreases with increasing atomic number A. It is very small for heavy elements. For heavier nuclei, partial energy transfers take place [15]. If the neutron energy is sufficiently high, inelastic scattering can also take place. For neutrons, the intensity equation becomes

$$I = I_0 \cdot e^{-\Sigma_{tot} L}$$

where $\Sigma_{tot} = \Sigma_{scatter} + \Sigma_{rad.capt.} + \ldots$ is the total macroscopic cross-section, the sum of the individual reaction cross-sections. The neutron free path is $\lambda = 1/\Sigma_{tot}$. In solid materials, λ for slow neutrons may be in the order of magnitude of a centimetre or less, while it is normally in the range of tens of centimetres for fast neutrons [15]. Because of this, neutrons are more likely to escape absorbing material in a warhead than gamma-rays. Although neutron emission from fission process is smaller than gamma-ray emission, neutrons might be more detectable in passive measurements.

While high-Z materials are not very relevant in shielding neutrons, other nuclides likely to be present in warheads can have a significant effect. Beryllium for example can be used in nuclear weapons as neutron reflector. Beryllium is ideal because it has a low atomic weight and therefore a large energy transfer in elastic scattering. In fact it has the largest macroscopic elastic scattering cross-section of all elements due in part to its high packing density. This could shield neutrons emitted from the fissile material. Furthermore, neutrons could be shielded by the conventional high explosives which are composed of low atomic weight molecules.

In order to evade problems due to unknown shielding, authentication steps have been developed, that rely less on gamma and neutron measurements performed directly at the weapons before dismantlement.

6. Authentication steps

A different verification mechanism is proposed that overcomes these problems (related to shielding of radiation coming from the fissile core which is an unknown parameter as explained above) to great extent while still ensuring a chain-of-custody:

The actual dismantlement of a nuclear weapon (after it has been removed from the carrier system and transported in a sealed and ID-tagged container) happens in a facility specifically designed for this purpose. Inspectors verify that the facility has just one (or a known number of) exit(s) and that no secret exits exist to ensure that no material can be transported in and out of the facility undetected by inspectors. The exit(s) are monitored by gamma and neutron detectors in order to detect any nuclear material entering or leaving the facility. In addition, inspectors enter the facility before the container with the warhead arrives to check that no additional (hidden) material is present. If for example fissile material is already present prior to dismantling the warhead, it could be presented as the actual warhead's fissile material, while the warhead is kept hidden without being dismantled.

While inspectors are present, the container with the warhead inside is brought into the facility, the seal and ID tag are verified. Behind the IB a number of measurements could already be performed. These could serve two main purposes: A very rough template could be recorded (e.g. very rough assessments of isotopic compositions or fissile masses, if possible behind the shielding) or rough attributes (e.g. the general presence of fissile material). Whether these measurements are possible without knowledge of shielding is a question that needs to be addressed.

After these measurements, the inspectors leave the facility so that the weapon can be removed from the container and be dismantled. During the dismantling process, the warhead will be separated into several components that will be put into separate containers. One container will contain the original fissile material without any shielding. Another container will contain the conventional high explosives. Other containers contain additional material present in the previous nuclear warhead.

Once the containers are closed, the inspectors enter the facility again. Now detailed gamma and neutron measurements of the separated components take place (behind an IB) which should in the end authenticate the nuclear weapon as a whole. The advantage of this authentication after the actual dismantlement is that only known shielding (i.e. the container) is present so that quantitative attributes can be assessed.

In the end, the IB can compare end results with the measurements taken at the beginning at the actual (not yet) dismantled warhead to create confidence that the material in the various containers after the dismantlement came indeed from the nuclear weapon so that the authentication of the components authenticate the nuclear weapon as a whole. If this is possible because of the shielding has not been determined yet.

While this entire process happens behind an IB, he/she needs to know the fissile material mass, which will most likely be one of the attributes during the authentication. This is necessary because in further verification measures within the chain-of-custody, inspectors need to be sure that all fissile material is vitrified or used for civil purposes and that none can be diverted. Fissile material shall be declared (including its mass) and monitored. Fissile material mass of a single warhead could, however, be considered sensitive information. Therefore an IB should not declare a single warhead's fissile mass, but the total fissile mass of a number of warheads instead. This way the average mass, but not individual masses of warheads can be determined, as long as different types of warheads were dismantled. Further required information would be the average isotopic composition of the fissile masses which will also very likely be a measured attribute.

7. Possible measurements for attribute determination

This chapter deals with defining the attributes to be measured. This list of attributes will not be exhaustive and more attributes than presented here will be necessary. This is however a list of what the authors consider the most important attributes: For both uranium and plutonium warheads, the presence and mass of fissile material should be determined, since a threshold mass is required to reach criticality. Also, the fissile material should be a pure metal, with only a minimum allowable content of oxides Significant presence of oxides would increase the critical mass and will increase the spontaneous fission rate through (α,n) reactions which is not wanted and will most likely not occur in nuclear weapons. By introducing this attribute, dummies with nuclear fuel (containing oxides) will be detected. For plutonium, the age can be determined as dates of plutonium separation might be known. The isotopic ratio between Pu-239 and other plutonium isotopes should be determined and exceed a certain threshold. For uranium, the degree of enrichment should exceed a certain threshold. A very strong indication of a nuclear warhead is both the presence of fissile material and high explosives.

The defined attributes need to be measured, therefore measurement techniques are presented for each attribute. Often, more measurement techniques will be possible that are not all listed because this paper's scope is only to present some potential possibilities of attribute measurements; sometimes it is not clear yet if a measurement technique can work. This will be studied in the present work. Although non-nuclear measurement techniques could be helpful, the study will be limited to nuclear non-destructive assay (NDA) using gamma and neutron detectors. A summary is given in Tables 1, 2 and 3.

attribute	*measurement system*	*measurement technique*
presence	gamma spectrometry	Pu-239 peaks at 345.0, 645.0 and 658.9 keV
age	gamma spectrometry	332.4 keV / 335.4 keV ratio
pure metal	gamma spectrometry	870.7 keV peak from O-17 de-excitation absent
isotopic ratio	gamma spectrometry	642.5 keV (Pu-240) / 646.0 keV (Pu-239) ratio
fissile mass	passive neutron multiplicity counting	spontaneous fission rate (Pu-238, Pu-240, Pu-242)

Table 1: attributes and measurement techniques for plutonium

attribute	*measurement system*	*measurement technique*
presence	gamma spectrometry	185.72 keV (U-235) and/or 1001.03 keV (U-238) peaks
fissile mass	active neutron multiplicity counting	induced fission rate (U-235)
pure metal	neutron-initiated gamma spectrometry	6129 keV gamma peak produced by 14 MeV neutrons
isotopic ratio	gamma spectrometry	185.72 keV (U-235) / 1001.03 keV (U-238) ratio after intrinsic self-calibration
	active neutron multiplicity counting	ratio of induced fission (U-235) to total transmission (U-235+U-238)

Table 2: attributes and measurement techniques for uranium.

attribute	*measurement system*	*measurement technique*
presence	gamma spectrometry	prompt gamma ray neutron activation analysis,measuring N presence and ratios of N/C, H/C and O/C, described in [16, 17]

Table 3: attributes and measurement techniques for high explosives.

7.1. Neutron coincidence and multiplicity counting

While gamma spectroscopy is widely applied and does not need to be described, this section gives a short overview over both passive and active neutron coincidence and multiplicity counting techniques. Neutron coincidence counting is used to measure coincident neutrons. This can be used to distinguish between reactions with only one neutron as a product and those with multiple neutron emissions. In our case, fission events, where usually multiple neutrons are emitted (with a certain neutron multiplicity distribution) should be detected, while in particular (α, n) and other uncorrelated sources should not be measured. Information regarding the features of coincidence counters is found in [14].

Alpha particles can produce neutrons through (α, n) reactions. This can become significant when isotopes with high alpha decay rates (e.g. U-233, U-234, Pu-238 or Am-241) are present [14]. The range of alpha particles in uranium and plutonium dioxide is roughly 0.006 and 0.007 cm, respectively[2]. If oxygen is intimately mixed with the alpha emitting material, a (α, n) reaction may take place. While there should not be large amount of oxygen present in nuclear weapon pits, impurities with oxides are possible.

The neutron flux emitted by the sample is affected by a number of possibly unknown properties [18]:

1. fission rate (the goal of neutron coincidence/multiplicity counting)
2. sample self-multiplication / variation across the sample
3. (α, n) reaction rate
4. other properties can be eliminated by careful calibration and counter design or are small or constant as described in [18]

The response function in passive coincidence counting can in particular be perturbed by the effect of self-multiplication that takes place in plutonium and uranium samples [14]. There are two common internal sources for self-multiplication: One is induced fission in an isotope by a neutron emitted by a previous fission event; the other is fission induced by a neutron from a previous (α, n) reaction [14]. Multiplication depends in particular on the sample composition and geometry [19].

For neutron coincidence counting (where both single and double multiplicities are measured), calibration using reference materials is necessary in order to obtain the fission rate. This is necessary because two parameters are measured, but at least three important parameters need to be solved, see above. Usually, the self-multiplication is not determined which requires representative reference materials. This is hard to achieve in the case of warhead authentication, where warhead designs remain unknown, so that representative reference materials can usually not be obtained.

Passive neutron multiplicity counting can determine the fission rate without the need of representative reference materials. For N unknowns in the function, N measured parameters are necessary. Multiplicity counting uses a third parameter (triple multiplicity) so that three unknowns can be solved, including the multiplication. Initial determination of detector parameters can be done with a Cf-252 source alone. Multiplicity counting based on this calibration can be slightly biased because of a detector's different efficiencies between Cf-252 and Pu fission neutrons [20].

Active neutron coincidence or multiplicity counting (using an external source to induce fission so that induced fission is measured as opposed to spontaneous fission) is more complex than passive neutron coincidence or multiplicity counting because of one additional unknown parameter, the coupling [21]. Coupling is the interaction of the source neutrons with the assay sample. The fission rate depends both on the induced fission rate and the coupling. This additional parameter has to be determined, as explained in [21]. It has to be noted that there is only limited literature and field experience on active neutron multiplicity counting [21]. Notably more field experience exists for active neutron coincidence counting: The Nuclear Materials Identification System (NMIS) [22] incorporates

[2] This range has been computed using the Bragg-Kleeman rule [14]:

$$\text{range} = 0.00032 \cdot \sqrt{A} / (\text{density}(\text{g/cm}^3)) \cdot \text{range in air}$$

The range in air for plutonium and uranium is 3.7 and 3.2 cm, respectively.

this technology; however application of NMIS to unknown weapon designs seems rather complicated because of reference measurements that are not available.
In general, it must be noted that in the mathematical models yielding the fission rate, assumptions have been made (explained in detail in [18]) that might not be entirely valid in the case of warhead authentication. How large the bias through these assumptions is needs to be assessed and what possible solutions could be. In particular, it is assumed that the neutron detector efficiency and probability of fission are uniform over the sample volume. This is called the “point model” as it is equivalent to the statement that all neutrons are emitted at one point [18].

7.2. Measurements for plutonium attributes

From gamma spectrometry, the presence of plutonium, its age and isotopic ratio can be deduced. For these purposes, so-called Pu300, Pu600 and Pu900 systems were developed for the FMTTD [5] that analyze plutonium characteristics in certain energy regions. When comparing gamma ray count rates to deduce isotopic ratios, it is helpful if gamma rays that are compared have similar energies so that the energy-dependent attenuation of absorbing material can be assumed to be the same for the peaks. It is more difficult to compare peaks that have very different energies because of significantly different attenuation coefficients. The age of plutonium can be determined by looking at Pu-241 and its daughter nuclides U-237 and Am-241 that have peaks between 325 and 350 keV [2]. Freshly separated plutonium contains Pu-241, U-237 results from alpha-decay of Pu-241, Am-241 is the result of beta-decay. Both U-237 and Am-241 decay further to two identical states of Np-237 by gamma ray emission including two intense peaks in the 325 and 350 keV range. The Pu300 code resolves the Am-241 and U-237 peaks from the Pu-239 peaks in the region [2]. The gamma ray emissions can be seen in Fig. 2. The branching decay of Pu-241 causes the levels that emit the 332.4 and 335.4 keV gamma rays to be populated at different rates. These are a known function of time, which allows for age determination (see Fig. 2).

The isotopic ratio $^{240}Pu/^{239}Pu$ can be determined using the Pu600 code which looks at the 630-670 keV region. The ratio can be determined by examining the peak areas of the 642.5 keV Pu-240 peak and the 646.0 keV Pu-239 peak [2]. This can be seen in Fig. 3.

The general presence of plutonium follows from Pu300 and Pu600 analyses: Pu-239 peaks can be found at 345.0, 645.0 and 658.9 keV. A criterion for determining the presence of plutonium could be that the peaks exceed the underlying continuum (due to background) by at least five standard deviations.

Gamma ray spectroscopy can also be utilized to analyze the possible presence of PuO_2 which should not be present in significant amounts in weapons built with metal plutonium. The presence of oxides can be determined for example by the Pu900 code that can analyze an 870.7 keV peak that is present if oxide is present. It results from de-excitation of the first excited state of O-17. The excitation results from alpha particles from the decay of plutonium that interact with O-17 via coulomb excitation which is an inelastic $^{17}O(\alpha,\alpha')$ process [2]. Another process leading to the same gamma ray emission are alpha particle reactions with nitrogen impurities in the oxide, $^{14}N(\alpha, p)$ [2].

Passive neutron coincidence or multiplicity counting can be used to determine plutonium mass. The mathematics of neutron multiplicity counting that give the plutonium mass is presented in [18]. The method detects the correlated fast neutrons emitted as a result of spontaneous fission decays in mainly Pu-240, but also Pu-238 and Pu-242. The primary quantity determined in passive neutron coincidence or multiplicity counting is an effective amount of $^{240}Pu_{eff}$ and consequently m_{240eff}, which represents a weighted sum of the three isotopes. m_{240eff} is the mass of Pu-240 that would give the same coincidence response as that obtained from all the even isotopes in the actual sample [14]:

$$m_{240eff} = 2.52 \cdot m_{238} + m_{240} + 1.68 \cdot m_{242}$$

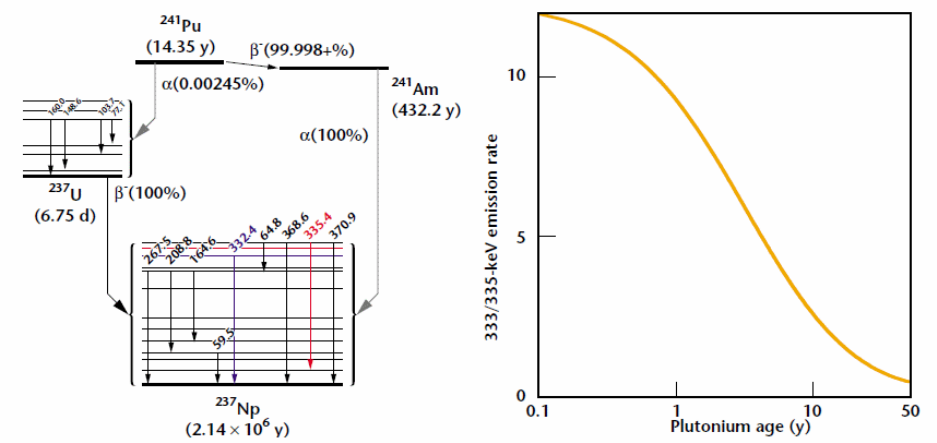

Figure 2: Left: The branching decay of Pu-241 shows the gamma ray transitions measured by the Pu300 method [2].

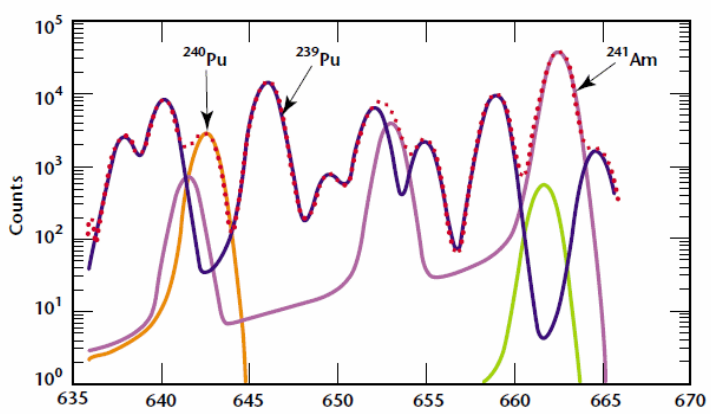

Figure 3: The 630-670 keV region of the gamma ray spectrum shows its resolution by nonlinear regression into its isotopic constituents for the Pu600 method [2]. The orange line corresponds to Pu-240, the light purple line to Am-241. The dotted line represents the total spectrum.

For the conversion of m_{240eff} into the total Pu mass, the isotopic mass fractions R_{238}, R_{240} and R_{242} must be known to calculate

$$^{240}Pu_{eff} = 2.52 \cdot R_{238} + R_{240} + 1.68 \cdot R_{242}$$

The total plutonium mass is then given as

$$m_{Pu} = \frac{m_{240eff}}{^{240}Pu_{eff}}$$

7.3. Measurements for uranium attributes

Measuring uranium attributes via gamma spectrometry is significantly more difficult because of the low gamma emissions rate. Therefore alternative measurement techniques are also suggested for attribute determination.

Presence of uranium could be determined by measuring the strongest gamma peaks: U-235 has its strongest peak at 185.72 keV, U-238 through its Pa-234m daughter at 1001.03 keV. The isotopic ratio would also be determined by comparing these two peaks. Since the attenuation of gamma radiation is, however, dependent on energy, these two peaks suffer from different attenuation coefficients, as their energies are not in the same region. The solution is to determine the ratio of the relative efficiency at the two energies by intrinsic self-determination. It can be shown that – for a series of gamma rays from a single isotope – the quotient of the photo peak counts at energy E_j^i and the branching ratio BR_j^i is proportional to the efficiency at energy E_j , $C(E_j^i)$ being the photo peak area of gamma ray j emitted from isotope i, N^i being the number of atoms of isotope i and $T_{1/2}^i$ being the half-life of isotope i [23]:

$$\frac{C(E_j^i)}{BR_j^i} \propto [\frac{N^i \ln 2}{T_{1/2}^i}] \cdot \varepsilon(E_j)$$

For this intrinsic self-calibration, gamma rays from several isotopes can be used as long as they have the same physical distribution [23]. Models exist to interpolate between gamma lines where the efficiency has been calculated [23]. In the case of uranium, intrinsic self-calibration can be done by comparing the peaks of the U-238 daughter nuclide Pa-234m, which also has a weak peak at 258.26 keV, which is close to U-235 peaks. Therefore, the isotopic ratio could also be calculated by comparing the 1001.03 keV (Pa-234m as indicator of U-238) with the 185.72 keV U-235 peak. However the spontaneous fission rate of U-235 is very small, it might be difficult to measure in reasonable time.

Therefore, a system using active neutron multiplicity counting might be more feasible. By adding an active neutron source, U-235 undergoes induced fission. In contrast, the total uranium (235+238) attenuates the transmission of active source neutrons through absorption and inelastic scattering. Therefore, the enrichment of uranium can be determined by measuring the rate of induced fission and the rate of direct source-neutron transmission [19]. The transmission is measured at the count distribution that corresponds to the time-of-flight of the source neutrons, as seen in Fig. 4.

The mass can also be obtained by active neutron multiplicity counting. The math works similar to determining plutonium mass as explained above, but coupling has to be incorporated.

To determine whether the fissile material exists as a pure metal or not, neutron induced gamma spectrometry can be used. A 6129 keV gamma line from oxygen can be observed by irradiating the fissile material with 14 MeV neutrons due to inelastic scattering, see Fig. 5. More information and further techniques are found in [19].

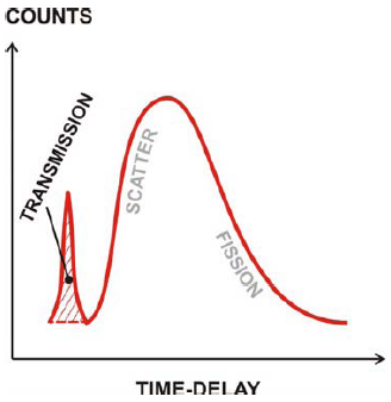

Figure 4: The integral of the first peak in the count-distribution is proportional to the neutron transmission rate and thus total uranium [19].

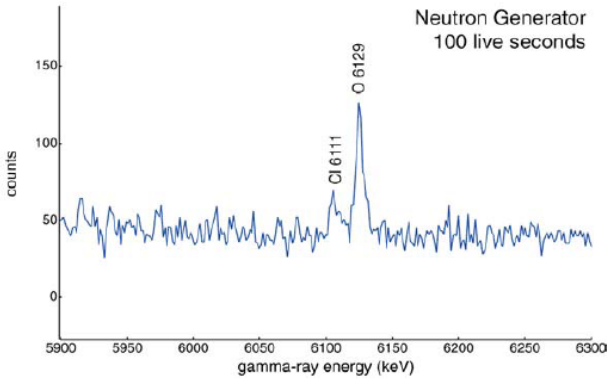

Figure 5: Gamma ray spectra for 14.1 MeV neutrons on oxygen, as obtained by A.J. Caffrey, INEEL, the sample was $COCl_2$ [19].

8. Conclusion and outlook

This paper has suggested warhead authentication as one element of the entire warhead dismantlement verification process. Information barriers, in particular the template and attribute approaches have been presented that allow warhead authentication without revealing sensitive information. Regarding the attribute approach, possible attributes have been presented including options to measure them. In the future, these options need to be validated. If possible, the following steps should be followed to validate the measurement options and build the IB:

1. Neutron and gamma signatures should be simulated for a variety of uranium and plutonium samples that differ in the relevant properties, especially isotopic composition, masses and geometry. Simulations can be performed by using Monte Carlo techniques. For neutron multiplicity analysis, the MCNP POLIMI code [24] can be used. The purpose of these simulations is to determine what signatures can be used to determine attributes and how the attribute determination depends on the signatures.
2. Experiments need to be performed with a couple of uranium and plutonium samples to compare the experimental results to the simulation results in order to assess the bias of the simulations. It is helpful if the samples could be to some degree representative of the items to be authenticated. It is obviously not possible for independent research to get access to fissile material used in nuclear weapons, and it needs to be assessed to what extent other samples that are available are useful for this task.
3. Knowing what attributes can be measured, the optimum set of attributes must be defined so that false positives and false negatives are minimized.
4. From the simulation and experimental results, an attribute algorithm needs to be written that uses the raw measurement data as an input and calculates an unclassified output using the attribute definitions.
5. Finally, such a system should be tested with unknown samples. As tests cannot be performed with nuclear weapon components, a “proof-of-concept” IB algorithm that uses different attributes that can be tested on non-classified forms of fissile material (e.g. reducing a threshold for minimal mass or enrichment degree) needs to be used.

9. References

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20 Novel technologies

Novel tools in support of future Safeguards implementation challenges

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Abstract:

One of the principle strategic objectives for the International Atomic Energy Agency (IAEA) is to deter the proliferation of nuclear weapons, by detecting early the misuse of nuclear material or technology, and by providing credible assurances that States are honouring their safeguards obligations. Early detection often requires sampling and analysis of materials associated with nuclear fuel cycle processes. To that end, the IAEA established a project to identify a broad range of suitable efficient and effective tools that can support the organisation's mission over coming decades.

There has recently been a huge increase in the availability of commercial-off-the-shelf (COTS) equipment targeted at identifying unknown substances in the field, based on a variety of physical techniques. Such equipment may prove beneficial to the IAEA nuclear safeguards inspector and provide them with more real time information when undertaking an inspection. In general, these products have been developed for the security/first responder market and thus, are usually already certified to standards similar to IAEA equipment standards, such as allowable operating and storage temperatures; protection against shock/vibration; shielding from RF interference etc. As such, the purchase and deployment of appropriate COTS equipment may represent a cost effective solution to improve the effectiveness and efficiency of the inspection process.

To best meet the needs of the end-user the majority of these products are targeted at the detection and identification of substances such as explosives, drugs and toxic industrial chemicals/materials. Numerous open-source reports exist on the efficacy of these instruments, which have been extensively tested by independent and government laboratories; however, very little data exist on the responses of these products when challenged with materials of interest to safeguards. Such basic questions such as the sensitivity, selectivity and dynamic range largely remain unanswered.

In order to begin to answer some of these questions, the IAEA recently purchased a COTS instrument based on the technique of Raman scattering. Initial evaluations of this instrument have concentrated on establishing a baseline of detection for several relevant compounds and the ergonomic factors that will define the deployment scenarios. This paper will highlight some of the results from this work.

Keywords: Portable; Raman, commercial; safeguards; inspector

1. Introduction

In meeting the need for in-field identification and/or verification of nuclear material; the IAEA safeguards inspector is well served with the choice of a large range of gamma and neutron detectors tailored to specific measurement circumstances. These instruments focus mainly on the measurement of uranium and plutonium content in declared material. However, there are other materials associated with various nuclear fuel cycle processes that could indicate clandestine activity early in the development and establishment phases and before the appearance of fissile products.Such materials may include, for instance,chemicals used in reprocessing or fluorine resistant lubricants. The IAEA is

enhancing its capabilities with the introduction of appropriate equipment for the in-field identification of such materials.

A large number of techniques have the technical capability for the detection and identification of safeguards relevant materials. These techniques include: high-performance liquid chromatography combined with mass spectrometry (HPLC/MS), gas chromatography combined with mass spectrometry (GC/MS), ion mobility spectroscopy (IMS), molecularly imprinted polymers (MIP), surface acoustic wave devices (SAW) and optical detection methods such as Fourier transform infrared detection (FT-IR). Most of the aforementioned techniques are just starting to become available as ruggedized, field-deployable units. However, the above methods generally require the end-user (e.g. the safeguards inspector), as well as parts of the instrument, to come in contact with the material. Such contact must be very carefully managed due to contamination issues for both the inspector and the instrument.

For safeguards implementation, it is often preferable for a measurement technique employed by an inspector to be both non-contact and non-destructive in nature. For this reason the use of Raman spectroscopy has been of great interest, especially with the recent remarkable improvements in the technique's portability.

Raman spectroscopy is named after the Indian physicist, Chandrasekhara Venkata Raman, who first described "a new radiation" (later known as the Raman effect) in 1928, for which he won the Nobel prize for physics in 1930. It is closely related to infrared (IR) spectroscopy, in that it records the vibrating, stretching, and bending movements of molecules. This allows the user to characterize an unknown liquid, solid or gas (in molecular form) by illuminating the sample with a low-power laser and matching the response to a ‘library’ of pre collected spectra.

The technique works by impinging monochromatic light on a target material. Much of the light passes through the sample unchanged,with some being absorbed, depending upon the wavelength of the light and the nature of the target material. A small fraction, about one tenth of one percent, is elastically scattered (this is known as Rayleigh scattering) and an even smaller fraction of the incident light (~ 1 photon in 10^6 or 10^7) may be scattered inelastically (Raman scattering). Raman spectroscopy probes the vibrational modes of the target molecules and it is those vibrational modes that can be regarded as a ‘fingerprint’ that uniquely identifies the substance.

2. Instrumentation Need

In identifying a suitable instrument a number of general characteristics were considered as being essential requirementsfor a commercial-of-the-shelf (COTS) system. For example, the main functional elements of the system must be integrated and fully contained within an independent handheld unit, which should include all the necessary hardware and software to undertake a measurement and provide a result e.g. excitation source, spectrometer, power source, electronics, computer processor and display. Because it was expected that the end-user would be carrying the instrument for relatively long periods of time, potential end-users requested that the system should be lightweight, with a total weight not exceeding 3 kg,with the batteries included.

In general, one of the main advantages of COTS systems is that the instrument is manufactured with environmental test standards in mind and, if certification is available to this effect, it represents a large cost saving to the IAEA in terms of not having to undertake such testing. Thus, it was requested that any system should be certified for environmental standards equivalent in nature to the International Electrotechnical Commission(IEC) environmental standards contained in the “IAEA Common Qualification Test Criteria for New SG Equipment” [1].

Because the end-user operates on a worldwide basis, it was important that the system should be transportable in civil carriers as “non-dangerous goods” and that it should have the capability of operating with various electrical grid supplies when recharging batteries or operating in a fixed mode.

It was requested that the system should automatically analyse the collected raw data and provide the end-user with an assessment of the type of material that has been targeted and allow for such measurements to be stored and transferred to a separate device for further analysis and archival.

In December 2010, the IAEA purchased a commercial Raman spectrometer, meeting the essential needs, as outlined above.The instrument is now undergoing further laboratory and field-testing as a part of the IAEA's instrumentation authorization process.

3. Authorization for Safeguards Inspection Use

Under the Safeguardsagreements between the IAEA and States based on INFCIRC/153, inspectors may make use of a number of technologies to draw safeguards conclusions including any "objective methods which have been demonstrated to be technically feasible" [2]. In order to demonstrate the technical feasibility of safeguards instrumentation, the IAEA has an established process of developing, evaluating and authorizing methods and instruments. Prospective instrumentation is usually placed in one of two 'categories', Category-C for instrumentation 'under development' or Category-B for instrumentation 'under evaluation', depending on the readiness level of the technology. Once the equipment has undergone testing relevant to its deployment scenario and conformed to expectations, it is elevated to Category-A (inspection use) equipment. This allows the instrument data to be used in drawing safeguards conclusions by the Agency.

The handheld Raman instrument has been placed directly into Category-B due to it being a commercial product with associated certification for safety and ruggedness. Throughout 2011 the instrument will be tested for its suitability for safeguards use. As part of this evaluation it will be necessary to test the instrument response against a range of compounds which have a relevance to safeguards.

The compounds of interest are listed in the IAEA *Physical Model*, whichanalysesthe known processes that are associated with nuclear fuel cycle activities and provides, inter alia, detailed descriptions of such things as the non-nuclear material used in the processes and by-products from these activities. The *Physical Model* weights each of these as strong, medium or weak indicators of the presence of a particular process.

Initial evaluations have focused on comparing the instrument's response to a range of materials available at the IAEA's Safeguards Analytical Laboratories,including such compounds as various uranium ore concentrates (UOC), ammonium diuranate (ADU), uranium trioxide (UO_3), uranium peroxide (UO_4) and various samples of 'yellowcake'. For each of these a 'library' scan was taken and stored on the instrument. In subsequent tests the instrument successfully identified all materials that had been added to the library. An example scan is shown below,in Figure 1, of a challenge with sulphuric acid (H_2SO_4) and the correct response from the instrument.

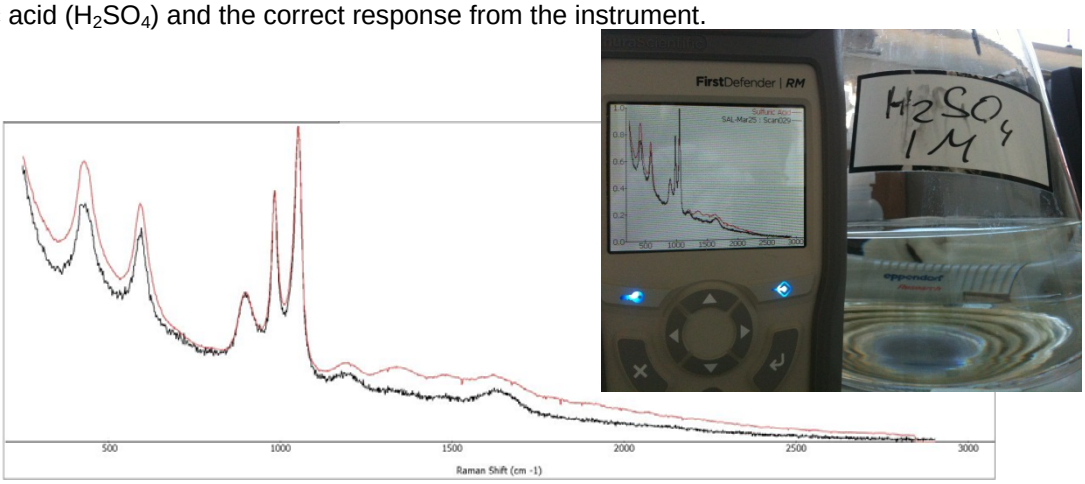

Figure 1: Example scan of a laboratory flask of sulphuric acid (black line) with the corresponding reference scan overlaid by the instrument (red line)

Future work will, inter alia, look at the addition of plutonium bearing compounds and also various acids and extraction agents that can be used fornuclear reprocessing.

Following the successful population of the instrument database with a range of materials of interest to safeguards, the instrument will go forward for field trails with end user. These field trials are intended to allow the IAEA to establish the necessary ‘Instrument Operating Procedures’ that will define the usage scenarios of the instrument. It is hoped that the handheld Raman instrument will enter category-A classification during 2012.

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LA-UR 11-02515

Application and Development of Laser Induced Breakdown Spectroscopy (LIBS) Instrumentation for International Safeguards

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Abstract

Advanced methodologies and improvements to current measurements techniques are needed to strengthen the effectiveness and efficiency of international safeguards[1]. The primary tool employed by the IAEA to detect undeclared processes and activities at special nuclear material facilities and sites still is environmental sampling. This type of environmental sampling is both time consuming and costly since many samples must be collected, packaged, and shipped to an analytical laboratory for analysis which in some cases can take weeks to months to complete. Los Alamos National Laboratory is currently investigating potential uses of LIBS for safeguards applications, including (1) a user-friendly man-portable LIBS system to characterize samples in real to near-real time (typical analysis time are on the order of minutes) across a wide range of elements in the periodic table from hydrogen up to heavy elements like plutonium and uranium, (2) a LIBS system that can be deployed in harsh environments such as hot cells and glove boxes providing relative compositional analysis of process streams for example ratios like Cm / Up and Cm / U, (3) an inspector field deployable system that can be used to analyze microscopic and single particle samples containing plutonium and uranium, and (4) a high resolution LIBS system that can be used to determine the isotopic composition of samples containing for example uranium, plutonium…etc.

In this paper, we will describe our current development and performance testing results for LIBS instrumentation both in a fixed lab and measurements in field deployable configurations.

Introduction

Laser Induced Breakdown Spectroscopy (LIBS) is a laser based optical method that can be used to determine the elemental composition of liquids, solids, and gases. In the LIBS technique, short pulses (typically 10 nanoseconds) from a laser are focused upon the surface of a sample where a micro-plasma is generated consisting of elements evolved from the surface and the gas above the surface. The emission from the plasma is wavelength resolved and detected using a dispersive device and a detector. The resulting spectrum is analyzed with a computer. The emission spectrum is characteristic of the emitting species in the plasma which are typically atoms, ions, and small molecules. If the spectra are collected and analyzed as a function of the chemical composition of the elements present, calibration curves can be generated from which semi to quantitative information can be determined. LIBS offers several advantages over classical wet chemical analysis techniques; (1) real-time or near real time

automated elemental analysis; (2) it is essentially non-destructive (only a few micrograms of material is removed from the sample per laser shot) with little or no sample preparation and handling required; (3) on-line or at-line analysis is possible, and ;(4) remote operation from multiple sites via fiber optics can be achieved. It is also a highly configurable technique meaning that instruments of many different shapes, sizes, and configurations can be designed, constructed, tested, and used to obtain chemical compositional information with varying levels of sensitivity, precision, and deployment (from fixed lab to field deployable systems).

LIBS Instrumentation

Laser Induced Breakdown Spectroscopy better known as LIBS, has been under development and applied to chemical analysis problems at Los Alamos National Laboratory and laboratories around the country and the world for over 40 years [2]. However, rapid development in LIBS was accelerated based primarily upon the pioneering work by Radziemnski, Cremers, and Loree at Los Alamos National Laboratory in the mid-nineteen eighties (1984) [3]. As an example of the maturity of LIBS technology, an instrument based on LIBS is scheduled for deployment to the planet Mars in 2011 for the elemental analysis of remote surfaces and features up to a remote measurement distance of 10 meters [4]. There are also national and international meetings devoted to improvements in and application of LIBS technology to chemical analysis problems [5].

Conceptually, the instrumentation for LIBS can range from simple to complex, depending upon the analytical analysis protocol and the level of precision and accuracy of the desired measurement. A schematic of a LIBS instrument is shown in Figure 1. In this diagram, the output typically from a Nd:YAG laser is focused onto the surface of a sample where a small plasma (typically a few millimeters

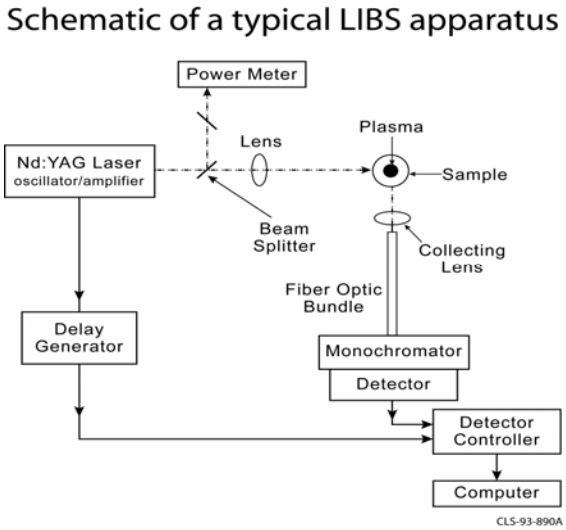

Figure1. Schematic of a typical LIBS experimental apparatus is shown.

in spatial dimension) is generated. Typically, the laser operates at 1064 nanometers with a pulse length of 7 – 10 nanoseconds. Depending upon the coupling of the laser light to the sample, a few to several hundred mili-joules of excitation energy is required to generate the plasma. The emission is collected with a lens and directed to a spectrometer using a fiber optic bundle. The emission is then analyzed by a computer.

Instrumentation Development and Performance Testing

Backpack Mounted Portable LIBS system

At Los Alamos National laboratory, a backpack mounted portable LIBS system has been developed and testing is in progress for the detection of the presence of actinides and other elements important to international safeguards. This system consist of a small Nd:YAG laser (Kigre, Inc) operating at 1/3 Hz with an output energy of 25 mj / pulse. The emission from the plasma is collected and directed to three Ocean Optic spectrometers (Model HR200+) using optical fibers. The spectra are detected with a CCD detector and analyzed with a small frame computer (Sony Inc.). The combined system weighs approximately 25 pounds, is completely self-contained, and operated in automatic mode using a battery. Currently the operational lifetime of the system is approximately 3.5 hours. A picture of the backpack LIBS system is shown in Figure 2. The technologist in the picture is Leon Lopez of the C-CDE group at Los Alamos National Laboratory. The green / silver unit at the end of the probe and near the wall is the sampling head that includes the small laser and focusing optics used to generate the plasma. This sampling head is also equipped with safety interlocks to protect the user. The green enclosure also prevents the user from coming in contact with dangerous stray reflections from the enclosed Class IV Nd:YAG laser. The black umbilical cord contains fiber optic cables for collecting emission from the plasma and directing it to the spectrometers and power cables for supplying power to the laser. A small form PC is located near Leon's right hand is the master controller for the laser, electronics, spectral collection, and data analysis. The electronic control unit is located in the backpack and contains the laser power supply, Ocean Optics spectrometers, and associated electronics for controlling the system.

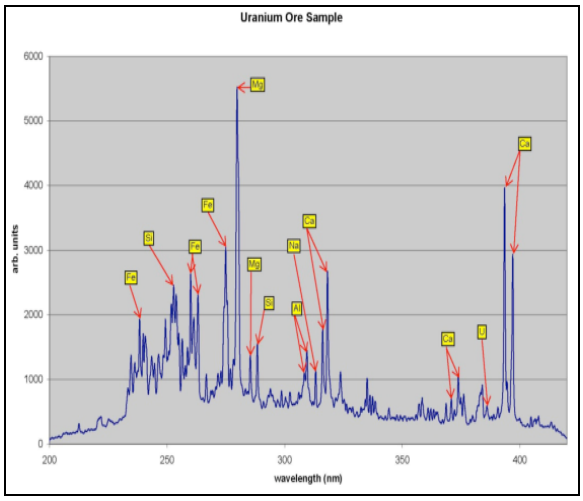

Figure2. A picture of the backpack system is being shown by Leon Lopez. On the right hand side is a typical LIBS spectrum of sample of depleted uranium in the region 200 – 420 nanometers (nm).

This system was used to analyze the following samples: (1) Magnets, AlNiCo, SmCo, and NdFeB; (2)Steels, 350 Marging steel, 250 marging steel, 304L SS, 316 SS, and A36 HRS (hot rolled steel), other steel alloys (carbon steel series 451-460); (3) Aluminum alloys, 6061 Al, 7075 Al, and 2024 Al; (4) Carbon fiber or graphite; (5) Aramid rubber; and (6) naturally abundance uranium in SRM 610 (standard reference material from NIST, Washington, D.C., USA) and uranium ore[6]. Altogether we analyzed 26 samples with a variety of matrices and chemical compositions. The concentration of uranium in the SRM and uranium ore samples was approximately 450 and 7500 ppm respectively.

We have identified approximately 30 analysis peaks or unique spectral signatures that can be used to detect the presence of uranium in environmental samples. The peaks that we have identified and assigned for uranium are listed in Table 1 where I and II refer to the neutral and first ionized excited electronic states of uranium atoms. Also we have identified and assigned unique spectral signatures for the magnets (30), aluminum alloys (40), and steel alloys (70). This set of data is similar to the data shown for uranium in Figure 2 above. Since each set has been analyzed for three spectral regions UV, VIS, and NIR each containing 2048 channels of spectral data, 6144 channels of data are then recorded per sample. The complete sample data set was placed in a validate database and then used to provide automatic sample identification for unknown test samples chosen at random from the combined data set without any prior knowledge of the identity of the sample under investigation. Using the algorithms and methodology developed, we correctly identified 24 out of 26 samples for a precision of approximately 92 percent. We are currently pursuing an expanded data set with an even wider range of chemical compositions and sample types.

Table1. Uranium peak Assignments from low resolution LIBS spectra.

Wavelength nm	Ionization State	Wavelength nm	Ionization State
268.37	U II	389.4	U II
270.63	U II	399.82	U II
277.00	U II	401.78	U II
278.44	U II	409.19	U II
295.63	U II	411.61	U II
302.22	U II	415.4	U II
310.24	U II	424.3	U II
311.16	U II	436.1	U I
339.47	U II	462.7	U II
350.76	U I	547.5	U II
353.4	U II	548.01	U II
367.01	U II	556.4	U II
385.9	U II	597.6	U I
387.4	U II	682.8	U I

Transparent automatic user friendly analytical analysis functionality has been integrated into this system. A view of this user friendly interface is shown in Figure 3 below.

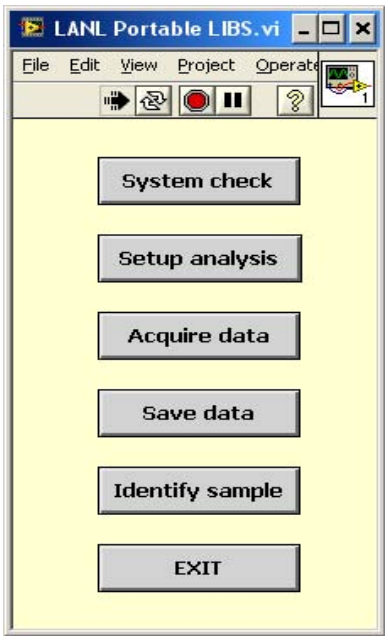

Figure 3. In this figure a transparent user friendly interface for controlling the backpack system, controlling and collecting data is shown.

The interface allow the user to: (1) perform a system check to verify that the system is operating correctly; (2) set the configuration for making a sample measurement; (3) acquire data; (4) save the data for further analysis; (5) perform sample identification by comparison to the sample data in the validated database; and finally exit or repeat the procedure for analyzing other samples. The transition and performance testing of this system from the laboratory to the field is in process. System improvements and testing will continue in the laboratory in parallel using a duplicate system. We also are in the process of developing a user manual and training for the safe and efficient use of the system. The intent is for the user to be safe and efficient in performing sample measurements with this system. To this end, we have also designed and installed appropriate safety interlocks to minimize or prevent the user from being exposed to Class IV invisible laser beams which can cause severe skin and eye damage.

Cart / Rack Mounted Field Deployable High Resolution LIBS System Development

We have designed, assembled, and testing is in progress for a high resolution LIBS system that includes an echelle spectrograph (LLA Instruments, Berlin, Germany). The spectrograph has a resolution of approximately 20,000 (wavelength / shift in wavelength). The emission is detected with an ICCD detector within the spectral range of 200 to 780 nm. The excitation source is a Quantel Nd:YAG laser operating at 20 Hz and with a 9 nanosecond pulse width and maximum output energy of 100 mj / pulse.

The system is controlled by an industrial computer operating on the windows XP platform. This system has the capability to be operated in one of three modes: (1) ***In situ*** with measurements distances of a few inches in a sampling chamber attached to a mobile platform; (2) remote measurements using direct optical access through the containment windows of hotcells or gloveboxes using a variable focusing head; and (3) remote measurements using fiber optic coupled probes at measurement distances up to approximately 100 meters both inside and outside hotcells and gloveboxes.

The remote functionality of this system in principle will allow monitoring and control of nuclear materials and processes at nuclear facilities in real to near-real time in a continuous and un-attended mode. Therefore any attempt to clandestinely remove or modify materials and nuclear facilities will be immediately detected. This system also can be used to provide isotopic and ratio analysis of samples of actinides (for example, isotopic measurements on samples of uranium, and important ratios that include U / Cm, Pu /, Cm, etc).

A prototype version of this system is shown in Figure 4. The picture on the left shows the sampling head (blue box mounted on a tripod) that contains the laser excitation source and optics for directing and focusing the laser beam through a window of a hotcell or glovebox. The sampling head also includes

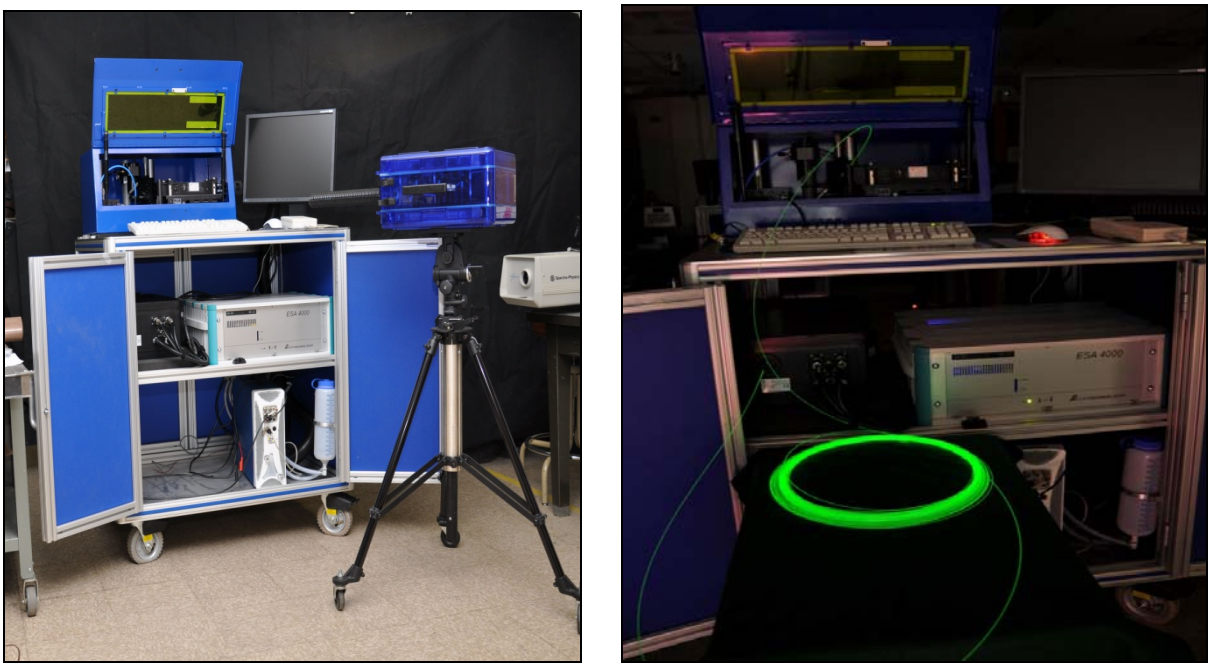

Figure 4. In this picture is shown a general view of the Cart / Rack mounted LIBS system on the left and the system coupled to a 50 meter fiber optic cable illuminated with a green alignment laser for visual effects on the right hand side.

optics for collecting the emission from the plasma and directing it to the spectrograph (black box to the left of the first level below the top of the platform) via a fiber optic cable. The blue box on the top of the platform with the access door open is the ***in situ*** sampling chamber. The light beige box also located on

the first shelf below the top is the industrial computer used to control the system. The vertical light colored box on the bottom shelf is the power supply for the Nd:YAG laser. The picture on the right side of Figure 4 shows the system coupled to a 50 meter fiber optic cable that was illuminated with a green alignment laser for visual effects. We have used this system to collect LIBS spectra through 2, 5, 20, and 50 lengths of fiber optic cables. A typical LIBS spectrum collected from a sample of depleted uranium is shown in Figure 5. A LIBS spectrum of thorium oxide in stearic acid binder is shown in Figure 6.

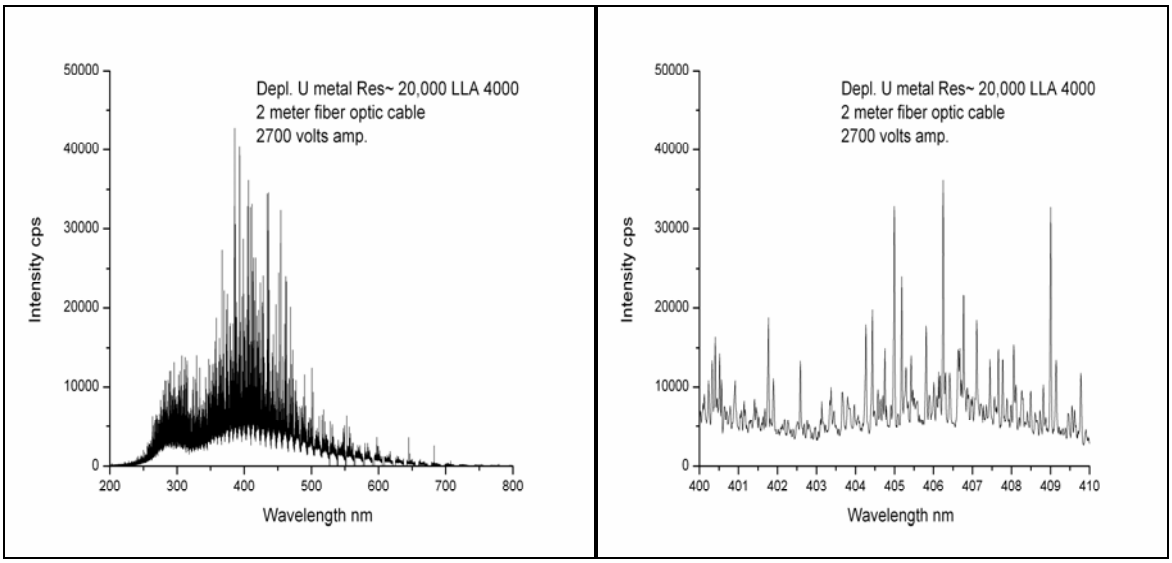

Figure5. In this figure we show a high resolution spectrum of a sample of depleted uranium between 200 and 750 nanometers. On the right hand side is shown a 10 nm section of the full spectrum.

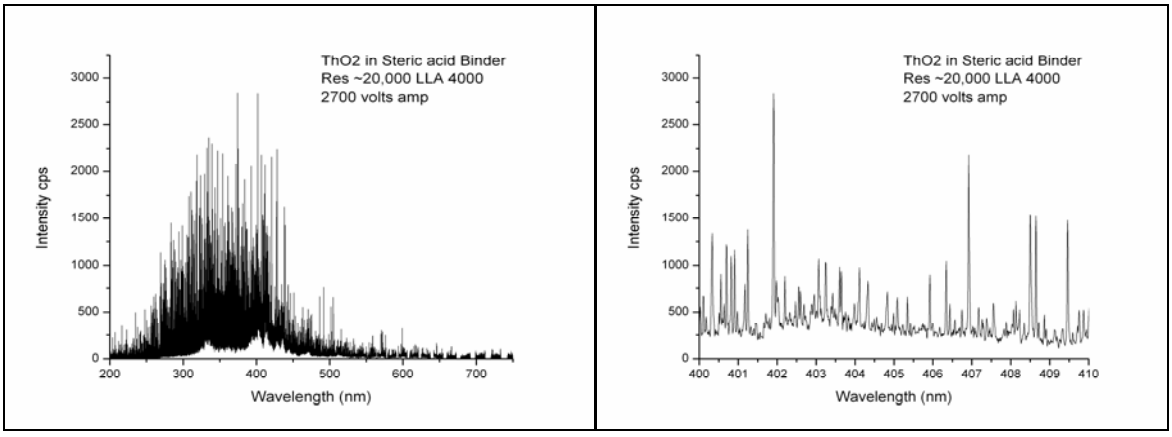

Figure 6. In this figure we show a full spectrum of thorium oxide on the left hand side between 200 and 780 nanometers. An expanded view of a 10 nanometer section is shown on the right hand side of the figure.

A careful and detailed review of the data shown in Figures 5 and 6 indicate that this type of spectra can be used to perform actinide ratio measurements on samples containing mixed actinides with a 20,000

resolution echelle based spectrograph. By contrast, it would be very difficult to use the low resolution spectra shown in Figure 2 (spectrum of a sample of depleted uranium), acquired with an Ocean Optics spectrometer to perform elemental ratio analysis of complex elements like the actinides.

High Resolution LIBS Isotopic System Development

We are developing an even higher resolution LIBS system for isotopic and ratio analysis for samples containing actinides. The core of this system is a high resolution echelle spectrograph with a resolution of 75,000 (wavelength / shift in wavelength). The resolution required to analyze enriched samples of uranium and plutonium is approximately 16,000 and 47,000 respectively. Thus this system can be used to perform isotopic analysis on samples of uranium and plutonium[7]. This is a much smaller compact spectrograph (approximately ¾ meter path length) compared to those used previously to perform isotopic measurements on samples of plutonium and uranium. For the plutonium measurements, a 2 meter scanning spectrograph operated in double pass mode was used. The uranium measurements were made with a 1 meter scanning spectrograph. The compact high resolution spectrograph along with an approximately 2 nanometer wide spectrum of a sample of depleted uranium is shown in Figure 7 below. The 242.3 nanometer line for uranium (the line to the right of the marked line) was used to perform isotopic analysis on samples of uranium (U-238 / U-235).

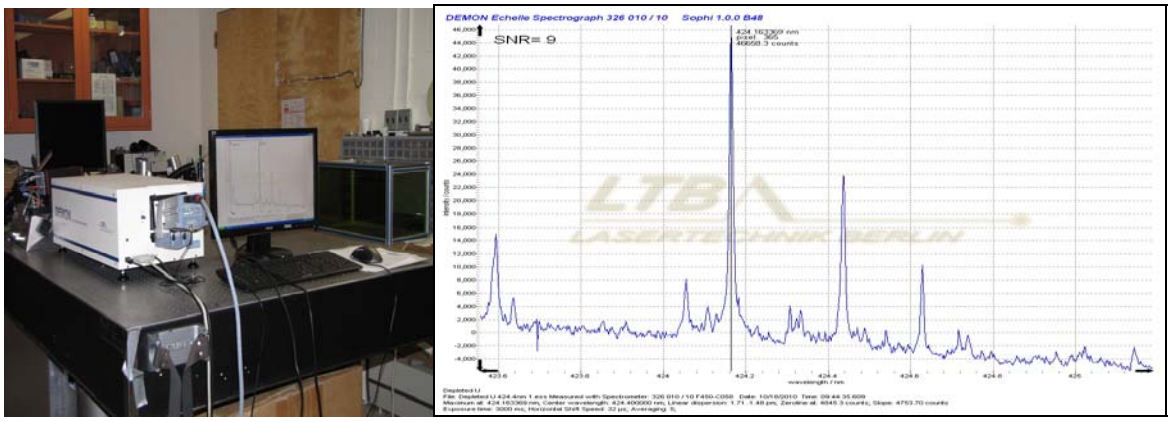

Figure 7. In this figure we show a high resolution spectrograph (75,000). A spectrum of an approximately 2 nanometer section for a depleted uranium sample is shown on the right hand side.

We hope to begin isotopic measurements on samples of uranium and plutonium here at Los Alamos National Laboratory soon using this high resolution LIBS spectrograph system.

Small Particle (approximately 100 micron in spatial dimension) LIBS Microscope Development

Finally, we are also developing a LIBS microscope system that can be used to analyze small particles of samples important to international safeguards. This tool can be used by inspectors that take swipe samples and want to make measurements in a field setting to determine if actinides elements are present. An image of this system along with a re-designed compact version is shown in Figure 8.

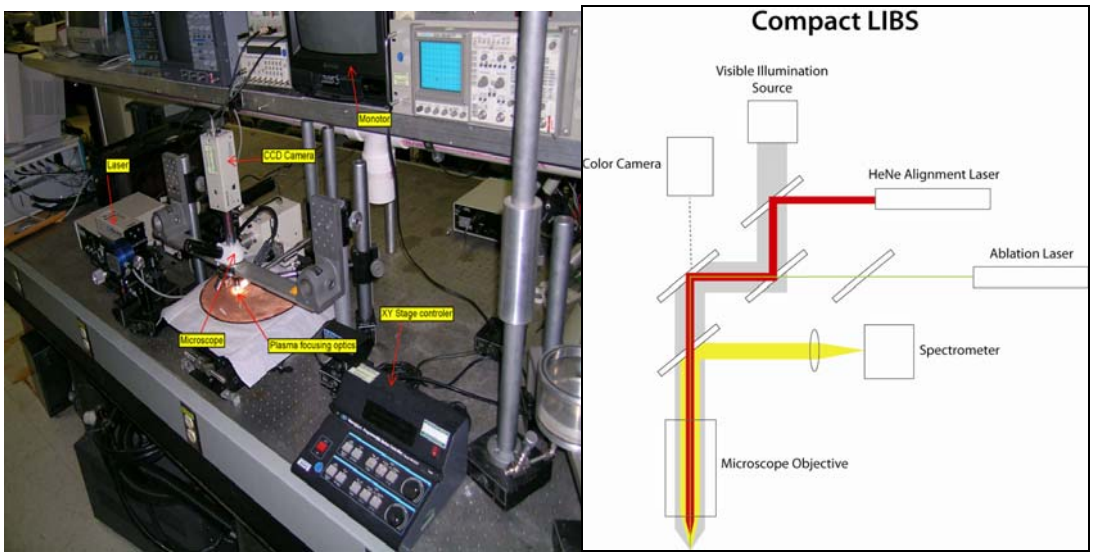

Figure 8. We show an image and a schematic of a compact version of the LIBS microscope in this figure.

This LIBS microscope can be used to analyze small particles on swipe media currently with a spatial resolution of approximately 100 microns. This tool has been used to analyze approximately 100 micron particles of aluminum and copper. A single particle LIBS spectrum of an aluminum particle is shown in Figure 9.

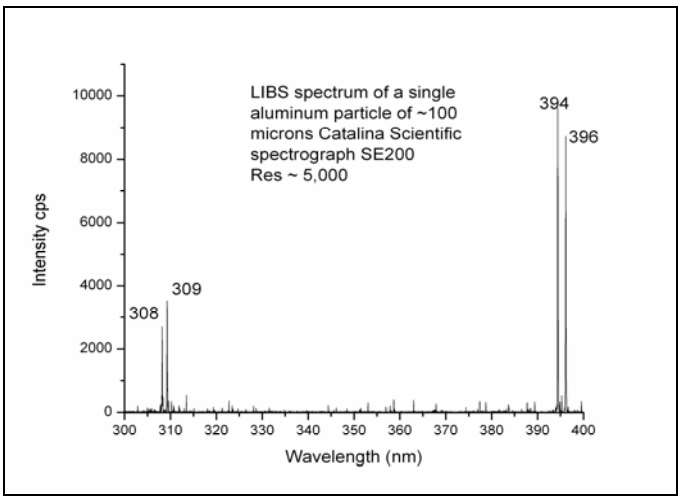

Figure 9. Shown is a LIBS spectrum of an approximately 100 micron particle of aluminum.

We are currently in the process of using this system to analyze small particles of depleted uranium and thorium. The results of this investigation will be the subject of future reports.

Conclusions

In this paper we have described some of our current development and performance testing results for LIBS systems designed to address the needs of the IAEA inspectors, the goals of DOE /NNSA's NGSI, and International Safeguards. The goals and needs will be supported by providing (1) improvements in the analysis times for special nuclear materials (typical analysis times on the order minutes can be achieved), (2) performing real-time process monitoring and control in nuclear facilities in a continuous and unattended mode, and (3) performing in-field, prescreening and analysis of environmental and nuclear material samples. All of the LIBS systems that we have developed can be deployed in a field setting thereby significantly reducing the number and therefore the cost associated with the collection, packaging, and shipping of samples for further analysis.

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The author would like to acknowledge support of this work from DOE/NNSA Office of Nuclear Safeguards and Security (NA-241). Los Alamos National Laboratory is operated by Los Alamos National Security, LLC for the United States Department of Energy under contract No. DE-AC52-06NA25396.

Simulation of Atmospheric Noble Gas Transport to Determine the Accuracy of Locating Unreported Reprocessing

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Abstract:

Verifying compliance with the Non-Proliferation Treaty (NPT) with regard to unreported materials and activities in nuclear facilities is one of the biggest challenges for nuclear safeguards. Novel Technologies such as environmental sampling that aim at the detection of proliferation indicators are being developed and improved.

The radioactive noble gas krypton-85 (^{85}Kr) can be used as a signature for undeclared nuclear activities as it is generated along with plutonium in the fuel cycle. Its chemical properties make ^{85}Kr an ideal tracer for plutonium separation activities. Improved sampling and measurement technologies in combination with atmospheric transport modelling can be used to detect and locate unreported reprocessing at distances of several hundred kilometres.
Atmospheric transport modelling (ATM) in forward mode from a suspected facility can help in deciding where to send inspectors for collecting air samples. Three regions have been chosen for case studies of simulated releases of ^{85}Kr using a Lagrangian Particle Dispersion Model. Utilising source-receptor sensitivities derived from ATM backwards calculations leads to the confinement of the possible source region (PSR). The results were analysed to determine the capability to accurately locate the PSR, depending on the number, position and sampling time of the employed measurement stations.
This information is used to determine cost effective sampling strategies and measurement methods to uncover a small, clandestine plutonium reprocessing campaign. "Catch the plume" scenarios using mobile sampling equipment are considered to monitor a region as well as temporary mini-networks and grab samples at facilities.

This paper is part of the project "Simulation of Atmospheric Noble Gas Concentrations to Assess Sampling Procedures for the Detection of Clandestine Reprocessing" (IAEA GER 1643).

Keywords: krypton-85; plutonium; reprocessing; ATM

1. Introduction

Verifying compliance with the Non-Proliferation Treaty (NPT) regarding unreported activities and undeclared facilities is a major challenge for nuclear safeguards. The discovery of clandestine nuclear operations in Iraq (1991) and Iran (2003) has led to the introduction of the NPT Additional Protocol (AP) which grants additional rights to safeguards inspectors and allows for new inspection strategies and techniques, such as environmental sampling [1]. Accordingly, new safeguards methods for detecting undeclared reprocessing facilities had to be identified [2].

One potential new signature for use in Safeguards applications is the noble gas ^{85}Kr which occurs as a fission product during reprocessing or reactor operation. Specifically, it is released during the dissolution and chopping process of spent fuel and a strong signature for verifying the operational status of a reprocessing facility as well as the detection unreported reprocessing activities. During the International Atomic Energy Agency's (IAEA) Technical Meeting (TM) on Noble Gas Monitoring Sampling and Analysis for

Safeguards Applications, 12-15 September 2005 in Vienna, a reference case for undeclared production and reprocessing of one significant quantity (SQ, 8 kg) of plutonium throughout one year was presented [3]. The experts participating the meeting provided an overview on existing noble gas collection and detection technologies, and analysed data available through open literature. The project "Simulation of Atmospheric Noble Gas Concentrations to Assess Sampling Procedures for the Detection of Clandestine Reprocessing" (IAEA GER 1643) takes up and improves upon these results to achieve an updated and more detailed understanding of the cost and capabilities of ^{85}Kr detection and its potential benefit to IAEA safeguards, taking into account recent advances and innovations in the field. Improving technological solutions for the detection capability of undeclared nuclear activities aims at strengthening the effectiveness.

2. Simulation of ^{85}Kr concentrations to determine noble gas sampling procedures for IAEA safeguards purposes

The radioactive noble gas ^{85}Kr is considered as a remote indicator for plutonium production [4]. Its chemical properties, long half-life (10.8 years) and the fact that it is a fission product and thus always generated along with plutonium in nuclear fuel make ^{85}Kr an ideal tracer for plutonium separation activities. Using state-of-the-art sampling and measurement technologies in combination with atmospheric transport modelling can help detecting and locating unreported reprocessing over long distances (>100 km) [5]. The first phase of task IAEA GER 1643 has demonstrated that outside Europe, approximately 70% of a single 3.2 TBq Kr release can still be detected 24 hours after emission (23 % within Europe), at a distance of up to several hundred km from the source depending on prevalent wind speeds [6]. After confirming the detectability of additional ^{85}Kr releases, the capability to locate the source region of the release has been investigated in the 2nd phase the task, the methodology and results of which will be described in the following sections.

The TM on Noble Gas Monitoring Sampling and Analysis for Safeguards Applications agreed on a *catch the plume* scenario for detecting and locating unreported reprocessing activities. In that scenario, the IAEA has a hint on a possible source region for noble gas emissions resulting from unreported activities. Since ^{85}Kr is diluted quickly in the atmosphere after its release, it is critical to obtain measurements in a timely manner.

Forward modelling can help identifying locations with high activity concentrations. Making use of complementary access under the AP, gives the IAEA the opportunity to send inspectors to the centre of the plume for taking air samples that can be either measured in field or be sent to a laboratory for analysis. By backtracking the measured activity concentrations, the possible release location can be estimated.

2.1. Source determination and localization using source-receptor sensitivities

Currently, no regional or world-wide measurement network for the detection of ^{85}Kr is operational and thus activity concentrations at fictitious sampling locations had to be simulated. The simulations were performed with the Lagrangian particle dispersion model FLEXPART [7]. FLEXPART is able to simulate transport, deposition, diffusion and radioactive decay of tracers forwards and backwards in time. The model has been applied forward in time to calculate the dispersion of ^{85}Kr from several release points and to determine fictitious activity concentrations at sampling locations (receptors). The backward mode of the model allows calculating source-receptor sensitivities (SRS) [8]. In this mode, particles are released from a receptor location (e.g. a sampling site) and a response function (sensitivity) to the emission input is determined.

The accuracy of the source localization capability depending on the number of sampling locations is of interest to Safeguards and has thus been analysed. For that purpose, the SRS fields have been calculated so that the concentrations c ($Bq\ m^{-3}$) for a specific radionuclide can be allocated to a possible source region. The correlation of the spatio-temporal source field S (Bq) and the corresponding SRS field M (m^{-3}) gives c at discreet locations (i,j) and time intervals n as follows:

$$c = M_{ijn}\, S_{ijn}. \qquad (1)$$

The SRS field M is a multi-dimensional array of multiplicators to translate a corresponding multidimensional array of sources into a concentration measurement at a receptor. As in a realistic scenario, it is likely to detect concentrations from the dispersed plume at

several sampling locations or concentrations one sampling location but for consecutive days. Hence the SRS concept (Eq. 1) is applied to a multiple measurement scenario with the activity concentration c_k ($Bq\ m^{-3}$) within a sample k:

$$c_k = M_{ijnk}\ S_{ijnk}. \quad (2)$$

The unknown in Equation (2) is the source term S_{ijnk} (Bq). It can be derived by solving the linear equation system, which is usually underdetermined. Strict regularization assumptions are thus necessary when inverting the matrix M_{ijnk} of the SRS field.
For this study, the regularization assumption has been applied that the source searched for is for an instantaneous grid cell. The problem can then be solved by the trial-and-error method. The possible source assumptions in space and time can be tested and the resulting hypothetical concentrations c_k can be compared with the actual measured concentrations (in our case: the simulated concentrations). The result is a correlation map of each possible source point for the measured (simulated) concentration for a given emission time. The correlation values on the map can then be interpreted as the relative probability of a release having occurred at a given time and location.
The analysis of the correlation maps for the performed case studies (see Section 3.1) provides information about the source localization capabilities of using acquired sample concentrations and atmospheric transport modelling.

3. Area of Interest for Safeguards

3.1. Simulated release scenarios and analysed sampling scenarios

To determine the capability of locating the possible source region, several ^{85}Kr releases have been simulated at three different release locations on the Northern Hemisphere, all at the same latitude. At each fictitious release point, releases have been calculated in the months of January, April, July and October to account for varying meteorological conditions throughout a year. The meteorological input data in a world-wide 0.5°x0.5° resolution comes from the European Centre for Medium-Range Weather Forecasts (ECMWF).
From each release point and for each of the release months, the emission and propagation of 1 PBq of ^{85}Kr has been analysed to find five different suitable receptors. 12 fictitious releases with equivalent emission parameters have been analysed.
To determine the accuracy of the source locatability depending on the number of sampling locations, for each of the three fictitious releases, five different sampling scenarios have been tested. Scenario 1 describes a measurement setup in which 8 consecutive 3 hour-samples are taken at one sampling location. For Scenario 2, two sampling locations are following this sampling pattern simultaneously, up to five sampling locations in Scenario 5. The agreement between the fictitious release point and the maximum correlation on the correlation map have been analysed and will be discussed in Section 3.2.

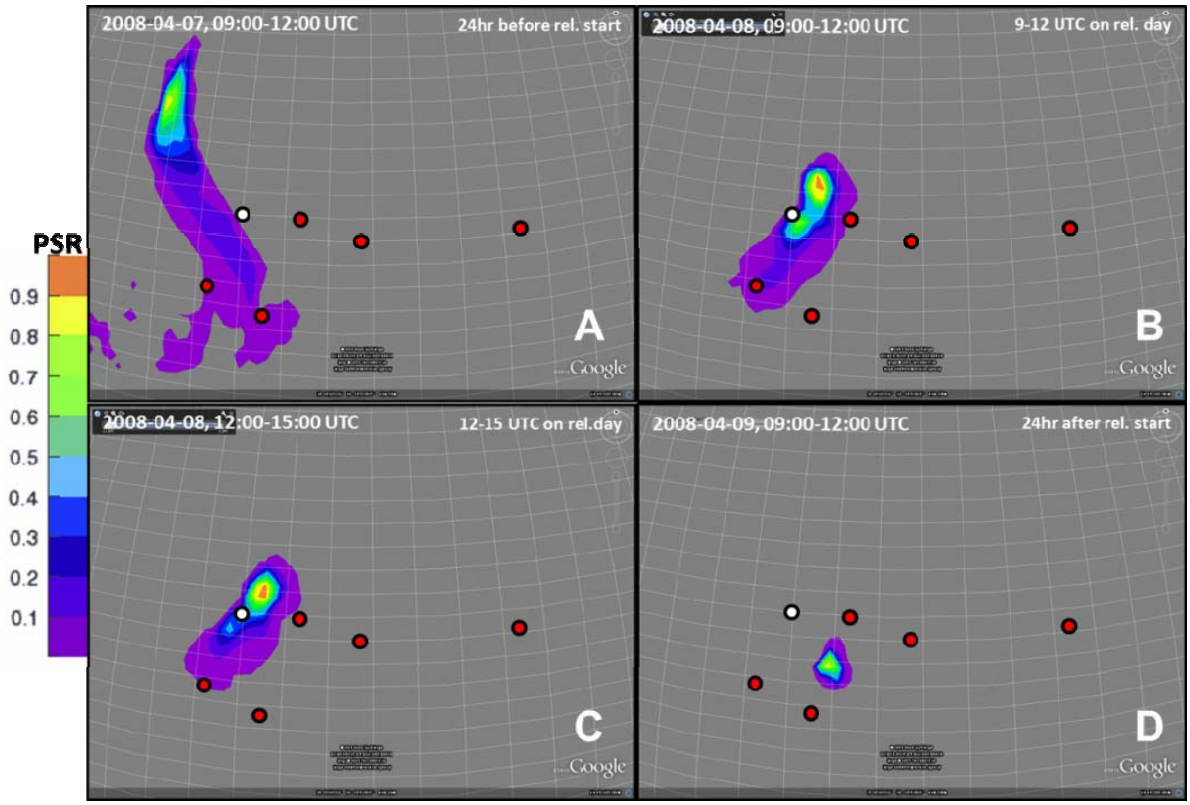
For determining and visualizing the correlation between the measurements (simulated sample concentrations) and the most likely release location and time, Web-Grape (Web connected graphics engine) has been used. Web-Grape is a heuristic tool to post-process SRS fields and calculate the correlation factor for a user-selected combination of SRS-data and sample concentrations.

3.2. Conclusion on the capability to estimate the possible source region using the catch the plume scenario

The catch the plume scenario assumes that hints reporting reprocessing activities are available. An IAEA analyst would then use forward modelling from the suspected area of reprocessing activities to estimate locations where the dispersed plume passes with the highest concentrations. Inspectors could be sent to various locations making use of Complementary Access under the AP. Different technologies for taking and measuring air samples are described in Section 4.1. The samples can either be analysed for their ^{85}Kr content on-site or be sent to a lab for analysis. The resulting activity concentrations can then be used for atmospheric backtracking.

The analysis of the simulated releases and the capability to determine the possible source region using five different sampling scenarios shows satisfactory results. 11 of the 12 releases were analysed, one release could not be analysed in backwards mode due to software problems. Thus statistical analysis was only performed on the remaining 11 releases. For each sampling scenario (see Section 3.1), the distance from the fictitious release point to the maximum correlation has

Figure 1: This figure shows the correlation between the stated sampling scenario (Scenario 5 in this case) and the most likely release point during 3-hourly intervals. The white bullet represents the fictitious release point, the red bullets mark the receptors which each has sampled 24 hours. Figure 1-A shows the correlation 24 hours before the release start, Figure 1-B and 1-C show the PSR during the first three hours and the last three hours of the release respectively.



been measured using Web-Grape. An example of such a correlation map is shown in Figure 1. The figure shows the correlation value (labelled PSR) between the stated sampling scenario and the most likely release point and time. The correlation map is shown for four different time stamps: a 3 hour interval, starting 24 hours before the fictitious release start (upper left), a 3 hour interval during the first three hours of the fictitious release (upper right), a three hour interval during the latter three hours of the fictitious release (lower left), and a 3 hour interval 24 hours after the fictitious release start (lower right). The figure shows the time dependence of the possible release regions.

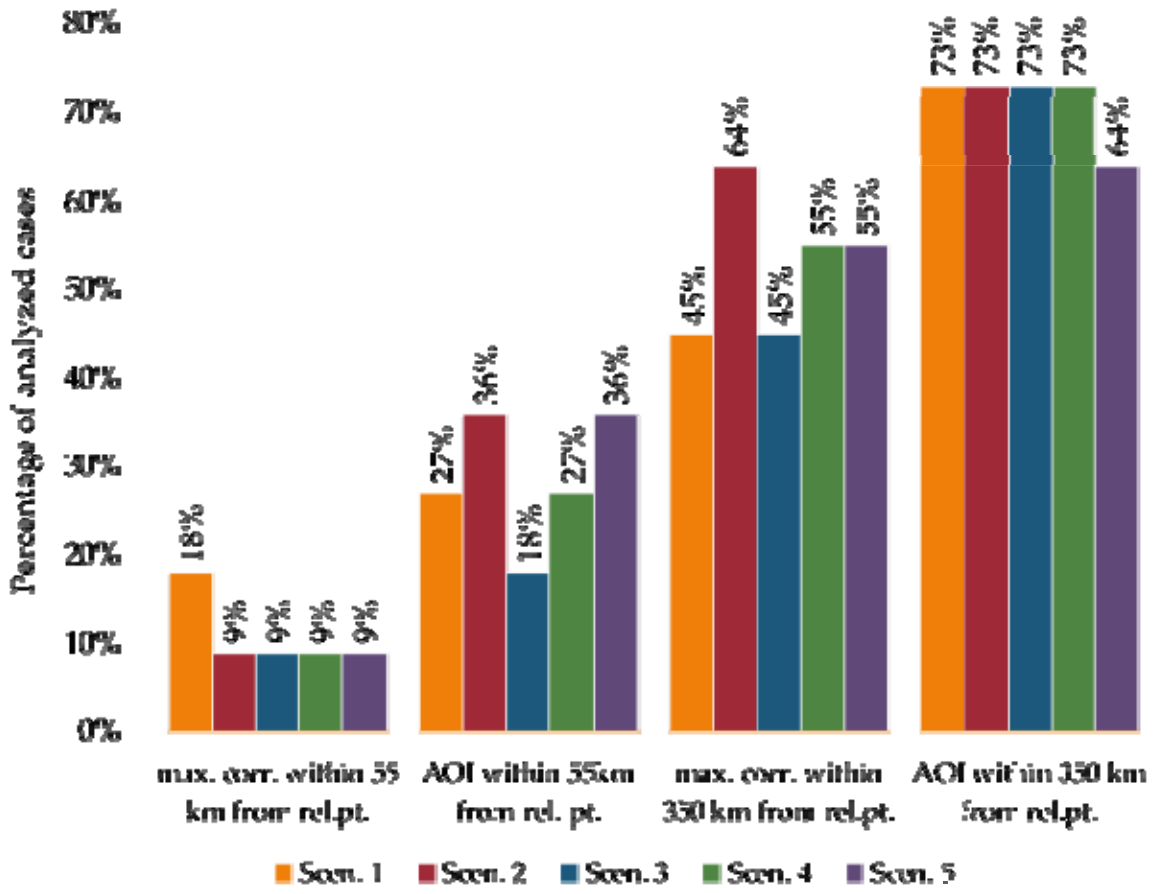
The distance from the fictitious release point to the point of maximum correlation has been determined for the four time stamps mentioned above.

As the area containing maximum correlation can be small and not be located directly at the suspected release point (due to uncertainties such as rapidly changing wind directions and growing correlation uncertainty when the sampling time is long after the release), the distance from the hypothetical emission location to the contour line of the so-called *area of interest* (AOI) has been determined.

The AOI is an area containing correlation values larger than 75% of the maximum correlation value found during the correlation map analysis for each sampling scenario.

To draw conclusions on the capability to determine the possible source region, the distance from the fictitious release point to the area containing maximum correlation values and to the contour line of the AOI have been measured for 55 cases in total (11 fictitious releases with 5 sampling scenarios each). Regarding the five different sampling scenarios, Scenario 2 (8 subsequent 3-hours samples at two different locations, see Section 3.1) is the most satisfactory compromise of source localization capability and sampling effort. In 9% of the analysed cases for sampling Scenario 2 the distance from the fictitious release point to the area containing the maximum correlation is within the error margin of the simulation. The resolution is 0.5° and defines the error margin given by the resolution which corresponds to 55km. In 36% of the cases, the AOI could be found to be within 55 km from the hypothetical release point for Scenario 2. Scenario 3, i.e. adding

Figure 2: This figure shows the percentage of analysed cases in which the correlation maximum and the AOI are in reach of the release point.



another sampling station and accordingly another set of eight 3-hours samples does not improve the source locatability. Figure 2 shows the statistical analysis and illustrates the percentage of analysed cases in which the correlation maximum and the AOI are in reach of the release point. The worsening of the source localization capabilities could be due to the fact that the third and following sampling locations are at larger distances to the fictitious release point and thus receive a more diluted activity concentration than the first two sampling stations.

In this study, source localization capabilities applying atmospheric backwards modelling have been studied systematically for more than one emission scenario. As of now, this has only been done for single release cases. Becker et al. (2007) [9] analyses intensively one release scenario and its source localization and is thus being presented here as a reference.

The study of Becker et al. (2007) analyses the ability to locate an unknown source location with a multi-model ensemble dispersion modelling (EDM). Twelve institutions, each operating their LPDM used for receptor-oriented backward atmospheric dispersion calculations, joined the study for that paper. The paper shows that the possible source region estimation of the ensemble-averaged EDM yields a backtracking result with a spatial error of 374 km corresponding to a deviation from the real source location.

Hence, the number of cases in which the maximum correlation and the AOI could be found within 350km range from the fictitious release point has been determined. The results of both statistical analyses, i.e. the number of cases in which the maximum correlation and the AOI are within 55km and 350km range of the fictitious release point, is shown in Figure 2. It becomes obvious that sampling Scenario 2 (two sampling locations, each sampling for 24 hours in 3-hourly intervals) gives the best source region estimation regarding the sampling efforts.

In 64% of the analysed cases the maximum correlation can be found within 350 km from the hypothetical release point for Scenario 2. In 73% of the cases, the AOI is in a range of 350 km from the fictitious release point. This is consistent for all the cases except sampling

Scenario 5, whereas the fifth sampling location is far away from the suspected and fictitious release point and thus only obtains a highly diluted signal.

Considering the sampling efforts, even Scenario 1 (one sampling location that samples 24 hours in 3-hourly intervals) yields satisfactory results for the source localization capability. In 73% of the analysed cases, the AOI can be found within 350 km from the release point with only one sampling location. In 45% the maximum correlation for the source region is in a range of 350 km from the fictitious release point for sampling Scenario 1.

The capability to estimate the source region is equally satisfactory with sampling Scenario 5; in 55% of the cases, the maximum correlation lies within a range of 350 km from the fictitious release point and in 64% of the cases, the AOI can be found within the same range. Nevertheless, the additional effort and cost suggest preferring Scenario 1 over Scenario 5.

Comparing the average distance from the AOI to the release point in this study and the spatial error of the source localization of Becker et al. [9], the results are satisfactory. The average distance from the fictitious release point to the AOI measures 396 ± 387 km for all analysed time stamps for Scenario 2.
Regarding sampling Scenario 2 during the fictitious release duration only (from 9:00 to 15:00 UTC on the 8th of the analysed months), the AOI could even be found at an average of only 267 ± 283 km from the fictitious release. For Scenario 2, the average distance from the fictitious release point to the region containing the maximum correlation is in the same range as the just quoted spatial error of the comparative study, which is a satisfactory outcome.
The size of the AOI has been determined for Scenario 3 (three sampling station, each taking 8 subsequent 3 hour samples). A reference to evaluate the size of the AOI is the size of the area for an on-site inspection for CTBT verification purposes. This area is to be determined by CTBTO for sending an on-site sampling team for verifying the conduct of a nuclear explosion. According to the CTBT, the area for an on-site inspection may not be bigger than 1,000 km², which corresponds to an area of about 32 km x 32 km, but the shape of this area may differ from a square [10]. The three smallest AOIs found in this study are 1,147 km² (24 km x 48 km, 1,606 km² (34 km x 47 km) and 7,159 km² (66 km x 108 km) large.

A small and precisely defined AOI is helpful in a case where the IAEA wants to send inspectors into a region for inspections. Compared to the size of an area for CTBTO on-site inspections, the size of the AOIs in this study is larger. Nevertheless, it should be kept in mind that CTBTO is using a multi-technology approach to determine this area. The analysis of seismic, hydro-acoustic, infra-sound and radionuclide data from a world-wide network and ATM are combined to get information on the area in which a nuclear test may have taken place. This study only used ATM for identifying the AOI, thus large areas are to be expected, especially when the receptors are far away from the suspected release point. Still, it is important to note that the IAEA can provide ways to minimize the size of the AOI by carefully evaluating the identified AOI. Areas such as oceans, lakes and rivers can be excluded from the AOI as no reprocessing would take place in such areas. Satellite imagery should be consulted as well, as it can help identifying areas without existing infrastructure, such as remote mountainous regions.
The remaining areas can be analysed again by using an increased resolution of the backwards simulations. The same measurements as for the preceding simulation can be used. Thus the AOI can be minimized according to the needs of IAEA Safeguards. Using the acquired samples for backwards simulations on a world-wide scale gives an overview of the possible source region. A more accurate specification of the release region can be achieved by increasing the resolution in the suspected area already identified and thus can improve the outcome of the source localization efforts.

4. Cost-efficiency and sampling strategies

A previous attempt to estimate the cost of supporting IAEA safeguards via ^{85}Kr measurement has been made [3]. That estimate was assembled ad hoc at a conference, based on the state of knowledge of 2005. Since then, the understanding of measurement and modelling techniques has vastly improved and warrants a new assessment of the cost involved.
A ^{85}Kr detection capability could be implemented into IAEA safeguards using a range of different approaches which can be generalised as three inspection strategies:

- Catch-the-plume
- Measurement network
- On-access grab sampling

Each of these sampling strategies could be implemented using different sampling and measurement techniques.

4.1 ^{85}Kr measurement techniques

Manual cryogenic sampling/measurement is the common method of processing ^{85}Kr samples, using a liquid nitrogen cooled cryogenic absorber in the field to extract the noble gas fraction from the sampled air. The krypton is obtained by heating the adsorber, separated using an activated charcoal column and measured using a thermal conductivity (TCD) and beta detector.

This procedure could be automated by constructing an **automatic cryogenic sampling/**device, much like the SAUNA and SPALAX monitoring equipment employed by the CTBTO for measuring radioxenon. Compared to xenon measurement, krypton measurement imposes additional technical difficulties, meaning that automated ^{85}Kr measurement is not readily available and would require a considerable development effort.

Atom trap trace analysis (ATTA) is a novel method for ^{85}Kr measurement suitable for very low sample sizes of about one litre of uncompressed air. This small sample size would facilitate ^{85}Kr sampling in the field, requiring no more than an evacuated gas bottle that is opened on site by the inspector.

4.2 Cost of ^{85}Kr measurement implementation

A cost comparison has been performed for all combinations of inspection strategy and measurement technique. All estimates include the required meteorological support and software.

Catch-the-plume is the least costly inspection strategy. As a base of calculations, it has been assumed that 3 bases around the globe are hosting measurement equipment to provide readiness for deployment within 24 hours of an inspection request, providing a detection capability analogous to Scenario 2 (see Section 3). Samples would be sent a central IAEA laboratory for measurement. The results are displayed in Figure 3.

In this scenario, the use of automated sampling and measurement equipment does not reduce the cost because of the small number of samples involved, but is associated with a much higher acquisition cost due to the development effort. ATTA compensates its relatively high acquisition cost due to the simplified sampling procedure.

A strategy using a permanent **measurement network** like the one included in the CTBTO's International Monitoring System (IMS) could only be realised using automated sampling equipment. Therefore, a cost comparison is not done in detail here.

The **on-access grab sampling** strategy mirrors the current practice of taking environmental swipe samples during safeguards inspections. Accordingly, an annual occurrence of 800 samples has been assumed for the calculation. Regardless of measurement technique, a single measurement device could process this amount of samples when operated around the clock, but two devices should be employed to allow for thorough maintenance.

Figure 3: The relative cost of implementing and supporting a catch-the-plume capability for IAEA safeguards. One campaign involves an overall 48 hours of sampling. Due to confidentiality issues, no actual numbers are given, but the arbitrary units are the same as in Figure 4.

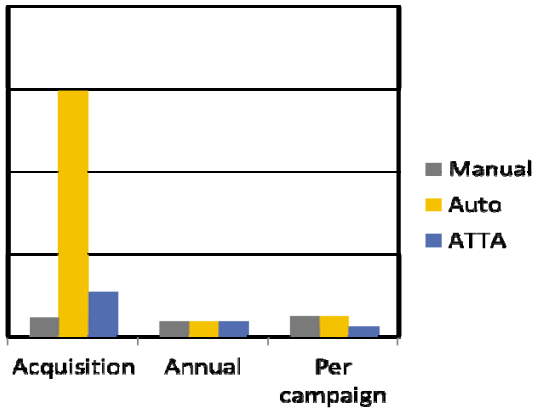
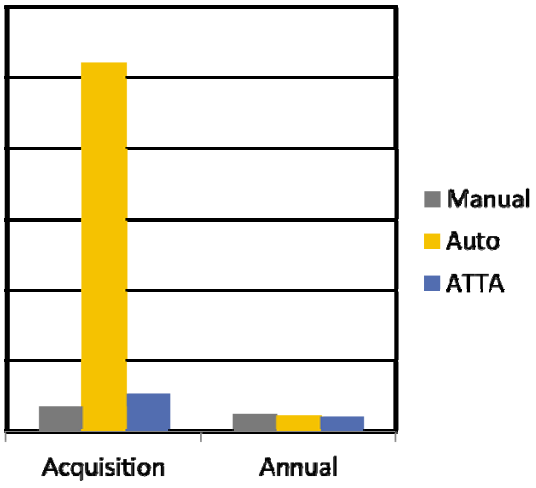


Figure 4: The relative cost of implementing and supporting an on-access inspection based 85Kr sampling capability for IAEA safeguards. 800 samples per year have been assumed. Due to confidentiality issues, no actual numbers are given, but the arbitrary units are the same as in Figure 3.



The costs are compared in Figure 4 using the same arbitrary unit as in the previous figure. Automated sampling requires a high investment due to the relatively high cost of an automated sampling device, whereas manual sampling is cheaper but unacceptably time consuming for the inspectors. Again, ATTA compensates the expensive measurement device due to the low cost of the sampling devices.

This cost comparison shows that ATTA will be the most suitable detection method for realistic safeguards ^{85}Kr measurement strategies, assuming it will be able to operate on small sampling sizes as planned. A fast, simple and ruggedized sampling procedure may prove crucial to enable a catch-the-plume approach or regular ^{85}Kr samples during safeguards.
In general, it can be assumed that the cost of utilizing ^{85}Kr in safeguards has been overestimated in previous reports.

5. Conclusion and Outlook

By simulating fictitious releases of ^{85}Kr corresponding to an illicit plutonium separation campaign, the project "Simulation of Atmospheric Noble Gas Concentrations to Assess Sampling Procedures for the Detection of Clandestine Reprocessing" (IAEA GER 1643) has demonstrated that source localization procedures can be successfully applied to ^{85}Kr monitoring. The resulting detection and localization capability is greater and more cost-efficient than suggested by earlier reports. Using only two mobile sampling devices, a 72 hour detection campaign can detect a ^{85}Kr source and narrow down its location to about 50–350km, even if only a small scale plutonium production campaign is assumed. Inspection capabilities will profit from the upcoming development of measurement techniques allowing for simple, lightweight sampling procedures such as atom trap trace analysis (ATTA).

6. Acknowledgements

The Authors would like to thank the German Support Program to the IAEA and the IAEA itself who funded this project under the Joint Programme on the Technical Development and Further Improvement of IAEA Safeguards between the Federal Republic of Germany and the IAEA. We also thank the German Foundation for Peace Research (DSF). Additionally, the authors thank the Central Institute for Meteorology and Geodynamics (ZAMG) for providing computational resources, necessary tools and work place for this research.

7. Disclaimer

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Reactor Neutrino Detection for Non Proliferation with the NUCIFER Experiment

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Abstract:

Neutrinos are the most abundant matter particles in the Universe. Thoroughly investigated in basic science, the neutrino field is now delivering first applications to the monitoring of nuclear reactors. The neutrinos are emitted in the decay chain of the fission products; therefore measuring their flux provides real-time information, directly related to the fission process occurring in the reactor core. Because of the very weak interaction of neutrinos with matter a neutrino detector can stand outside the core containment vessel and provide a non-intrusive and inherently tamper resistant measurement.

After a brief review of the existing data and worldwide projects, we present the NUCIFER experiment. The active part of the detector is a tank filled up with one ton of Gadolinium-doped liquid scintillator. 16 photomultiplier tubes, isolated from the liquid by an acrylic buffer, read out the light produced by the interaction of a neutrino with the protons of the liquid. The tank is surrounded by plastic scintillator plates to veto the cosmic rays. Then polyethylene and lead shielding suppress the background coming from external neutrons and gamma rays respectively.

The NUCIFER detector has been designed for an optimal compromise between the detection performances and the specifications of operation in a safeguards regime. Its global footprint is 2.8 m x 2.8 m and it can monitor remotely the nuclear power plant thermal power and Plutonium content with very little maintenance on years scale. The experiment is currently being installed at the OSIRIS research reactor (70 MW) in Saclay, France. First data are expected by autumn 2011.

This work is done in contact with the IAEA/SGTN division that is currently investigating the potentiality of neutrinos as a novel safeguards tool. A dedicated working group has been created in 2010 to coordinate the simulation effort of various reactor types as well as the development of dedicated detectors and define and eventually test the end product to be used by the agency.

Keywords: *neutrino; non-proliferation; safeguards; remote monitoring; reactor*

1. Introduction

Neutrinos are largely produced in reactor cores by β-decay of neutron rich fission products (FPs) from U and Pu into more stable nuclei: ${}^{A}_{Z}X \rightarrow {}^{A}_{Z-1}Y + e^{-} + \bar{\nu}_{e}$. Nuclear reactor power comes from the energy produced both during the fissions and the β-decays. This second contribution is about the 7%

of the total amount of produced power and it is called decay heat. The global released energy per fission is about 200 MeV with the emission of 6 antineutrinos. This means that the flux emitted by a 1 GW_{th} reactor is ~1.5 10^{20} antineutrinos/second. This huge emitted flux allows us to detect their signal with a relatively small detector (1 ton scale) placed a few 10 m from the core even if the interaction cross section between matter and neutrinos is very tiny (~10^{-43} cm^2).

The FPs' mass distribution depends on the fissioning isotopes (^{235}U, ^{238}U, ^{239}Pu and ^{241}Pu) and incident neutron energy. The mean energy released per fission, the mean number of neutrinos emitted by FP beta decays and their mean energy are different depending on the initial fissioning isotope. So the detection of these neutrinos would give a remote and real time image of the core composition.

In section 2 we will present the principle of reactor monitoring by neutrino detection and the world effort on this subject. In section 3 we will present our detector prototype, Nucifer, while In section 4 we will report on its expected performances.

2. Reactor monitoring by antineutrinos

2.1. The principle

Reactor antineutrinos are usually detected by using large volumes of liquid scintillator (hydrocarbon) doped with Gadolinium. The detection reaction is the inverse β-decay $\bar{\nu}_e + p \rightarrow e^+ + n$. The e^+ detection produces a prompt signal of energy $E_{prompt} = E_{\bar{\nu}_e} - \left(M_n - M_p\right) + m_e$, with M_n the neutron mass, M_p the proton mass, m_e the positron mass and $E_{\bar{\nu}_e}$ the neutrino energy. This first pulse is followed by a delayed (τ ~ 30 μs) signal induced by the radiative capture of the neutron on Gd with the emission of a gamma cascade of total energy ~8 MeV. The mass excess of the final state of the inverse β reaction implies a kinematical threshold of $(M_n–M_p+m_e)c^2$= 1.8 MeV for the antineutrino energy.

The two main fissile isotopes contained in the fuel of a pressurized water reactor (PWR) are ^{235}U and ^{239}Pu. Fresh Uranium fuel is typically enriched at 3.5% in ^{235}U, while ^{239}Pu is produced by neutron captures on the original ^{238}U followed by two consecutive β-decays: $^{239}U \rightarrow {}^{239}Np \rightarrow {}^{239}Pu$. During a reactor cycle ^{235}U is burned while ^{239}Pu is produced. This means that the relative contribution to the total number of fissions induced by these two isotopes changes over time: it increases for the ^{239}Pu while decreasing for the ^{235}U. This is called the “burn-up” effect.
Because the FPs of these two isotopes have different atomic masses their β-decays result in different neutrino fluxes. The idea of a neutrino probe to monitor the reactor fuel composition is based on the hypothesis that we are able to discriminate between different generated fluxes.

Fission parameters	^{235}U	^{239}Pu
E/fission[a] (MeV)	193.5	198.9
<Eν> above threshold (MeV)	2.94	2.84
Nν/fission above threshold	1.92	1.45
$<\nu_{int}>$ (10^{-43} cm^2)	3.20	2.76

Table 1. Relevant fission parameters of ^{235}U and ^{239}P.
[a] Only the energy contributing to the thermal power is quoted here (i.e. total released energy minus antineutrino energy)

Considering the key parameters for ^{235}U and ^{239}Pu fissions summarized in table 1, we can calculate the ratio of detected antineutrinos in the two extreme cases, where all fissions come from pure ^{235}U or pure ^{239}Pu to produce the same thermal power:

$$\frac{N_v^U}{N_v^{Pu}} = \frac{\left(\frac{N_v}{fission}\times\sigma\right)^U}{\left(\frac{N_v}{fission}\times\sigma\right)^{Pu}} \frac{\left(\frac{E}{fission}\right)^{Pu}}{\left(\frac{E}{fission}\right)^U} \approx 1.6$$

This large difference suggests the possibility to use the antineutrino rate to monitor changes in the relative amounts of ^{235}U and ^{239}Pu in the core.

The neutrino detection monitoring method relies on relative antineutrino flux measurements made before and after a given period of time (suspicious reactor stop, refuelling, ...). A neutrino flux change can be induced both by a change in the core composition or by a change in the produced thermal power. Assuming an independent knowledge of thermal power, the comparison between two relative neutrino flux measurements would allow us to calculate the relative change in U/Pu composition of the core. The big advantage of making relative measurements is that all normalization errors (solid angle, detection efficiency, …) cancel out.

Another monitoring method can be obtained from the declaration of the power history of the operator: the expected time evolution of the neutrino flux can be predicted and compared with the data. This controls the correct operation of the reactor over the course of the cycle [1].

2.2. World effort

The idea of applying neutrino to reactor remote control was firstly presented to the International Atomic Energy Agency (IAEA), the United Nations agency in charge of the development of peaceful use of atomic energy, in 2003, during a meeting with the neutrino scientific community. The IAEA asked for a feasibility study to determine whether antineutrino detection methods could provide practical safeguard tools for selected applications. This encouraged the international community to work on prototype design and performance studies. Two others meetings have been realised by the AIEA in October 2008 [2] and October 2010 [3]. Results by pioneering experiments like the SONGS1 detector [4], new projects like NUCIFER and prediction of non-proliferation monitoring scenario coming from the accurate simulation of different reactor types [5][6][7][8] were presented. During the last meeting the AIEA has created a dedicated working group on this last research developments [9].

This interest from the AIEA motivated the international neutrino scientific community in search for technical solutions for detectors to be installed close to nuclear reactors. The requirement is to build a relatively small neutrino detector, remote controlled, safe and easy to operate by non-experts. The main challenge is the detector placement close to the Earth surface. Neutrinos are weakly interacting particles and close to the Earth surface their signal can be easily mimicked by the cosmic ray induced signal. This is why, typically, neutrino detectors are place in deep underground laboratories. The non-proliferation specific requirement of compactness implies to be close to the reactor and quasi on Earth surface. The main challenge is, then, the neutrino signal extraction from the background induced by cosmic rays.

The pioneer experiment in antineutrino detection for reactor monitoring is the one performed in 1986 at the 1.3 GW_{th} Rovno reactor in Russia [10]. The detector was a $1m^3$ plexiglass tank filled with Gd-doped liquid scintillator and surrounded by 84 photomultipliers and was placed at about 18 m from the reactor core. With a daily detection rate of the order of 1000 events they were able to clearly see a ~6% burn-up effect on neutrino flux over a reactor cycle and to measure the proportionality law between neutrino rate and thermal power [11].

Some other results demonstrating the possibility of monitoring the antineutrino flux from nuclear reactors in correlation with burn-up effect and refuelling have been shown by the SONGS1 experiment developed by the Lawrence Livermore National Laboratory in the US. The target was 0.64 tons of Gd doped liquid scintillator and the detector was placed at 25 m from the core of the 3.46 GW_{th} San Onofre Nuclear Generating Station (SONGS), in California. With a mean number of ~400 detected neutrinos per day, this experiment allowed to measure the burn up effect over two reactor cycles

(about 18 months) and the ~10% step in rate due to the replacement of burnt fuel containing 250 kg of ^{239}Pu by fresh fuel containing 1.5 tons of ^{235}U [4] [12] .

These two detectors were placed underneath a significant amount of construction material allowing some shielding to cosmic rays. Prospects of future deployment closer to surface have triggered the study of different techniques to improve background rejection: standard doped liquid scintillator detectors like Nucifer in France or Kaska in Japan, Gd doped water at Angra in Brazil and organic scintillators layed with high neutron cross section materials at the LLNL in the US, or even new detection techniques like coherent antineutrino-nucleus scattering in Taiwan [9].

3. Nucifer experiment

3.1. Detector design

The Nucifer concept has been developed by following the IAEA requirements and the necessity of a good detection efficiency and a high background suppression. This last point is the real challenge for this detector since it will be placed close to the Earth surface, where the cosmic ray induced background (muons and induced secondaries like neutrons) becomes really important.

The detector design has been realized in collaboration by the CEA/IRFU/Spp and SPhN of Saclay and the CNRS/IN2P3/Subatech laboratory. The design was optimized using a dedicated GEANT4 [13] simulation of the detector response to signal and background and by performing several background measurements at the Osiris (CEA-Saclay) and ILL (Grenoble) research reactors where the detector will be first tested.

The two main background sources are cosmic muons (and induced particles like fast neutrons) and gammas generated by the reactor, since the detector will be placed a few meters away from the core. The first type of background can give rise to a correlated signal (proton recoil from neutron scattering which can mimic the positron prompt signal plus correlated neutron capture to reproduce the delayed signal), while the second type of background contributes to the accidental signal (gamma detection which mimic the prompt signal plus accidental neutron capture). The gamma spectrum and flux at the 2 research reactors have been measured and simulated for the Osiris reactor [14]. The maximal gamma energy is around 10 MeV.

The optimised detector is composed of a neutrino target detector, an active muon veto to tag the muon induced backgrounds and neutron and gamma shielding. The global view of Nucifer is reported in figure 1, while the target detector and one module of the muon veto are shown separately in figure 2.

The neutrino target detector is a steel cylindrical tank (height 1.8 m, diameter 1.2 m) with internal surface coated with white Teflon for compatibility with the scintillator liquid and to increase light diffusion. The tank is filled with about 0.85 m3 of Gd-doped liquid scintillator. A 25cm thick acrylic disk optically couples the PMTs with the liquid surface while shielding the intrinsic PMT radioactivity from the scintillator and ensuring a more uniform response in the whole target volume. 16 large PMTs (8 inches diameter) located at the top of the detector vessel ensure an efficient light collection. They are always kept in an inert Nitrogen atmosphere inside the vessel reducing further the fire hazard. A LED based light injection system allows to monitor the PMT gain and possible instrumental drifts. In addition small radioactive sources could be deployed along the target central axis inside a vertical tube.

Two hermetic layers of shielding protect the detector from gamma and neutron backgrounds. Starting from the outer layer we can find a 10 cm Pb layer and a 14 cm Polyethylene layer.

The muon veto detector is placed between the shielding and the neutrino target as shown in figure 1. This is mandatory in order to protect the veto from the high energy gamma flux coming from the reactor core. The veto will surround 5/6th of the neutrino target. It is based on a novel concept: 32 modular detectors, each one containing a 5 cm thick plastic scintillator of 150 to 170 cm length and 25 cm large. The scintillator is observed by one PMT decoupled from its surface. The scintillator thickness has been chose in order to discriminate cosmic muons from gamma background. The used

technique is a threshold principle to cut a maximum of gammas. Indeed cosmic ray muons are generally high-energy particles, so that we can assume that all are minimum ionizing. For plastic scintillator, the minimum ionizing value of dE/dx is ≈ 1.9 MeV.cm^{-2}. Therefore a 5cm plastic slabs lead to have an averaged deposited energy of about 10 MeV. Such a threshold can be applied to eliminate gammas that deposit under this threshold. Indeed the threshold value depends also on the light collection uniformity. The choice of one PMT per module has been made to respect the cheapness requirement of the project, but this clearly penalizes the uniformity of module response to light collection. To optimise the module geometry and PMT positioning for maximal uniformity response and to study the detector performance we developed two simulations based on GEANT4 and Litrani [15] codes. The final measured detection non-uniformity is less than a factor 2.5. The threshold of each module has been set to minimize the gamma detection while maximizing the muon detection efficiency to be greater than 95%. Energy threshold calibration and detector performance have been studied by performing several measurements on one prototype module: muon signal detection, measures with gamma sources (gamma prompt from AmBe, ^{60}Co) and a campaign of measure at the Osiris reactor where the detector will be placed. The final estimated threshold in energy is between 2 MeV and 5 MeV depending on the event position inside the scintillator.

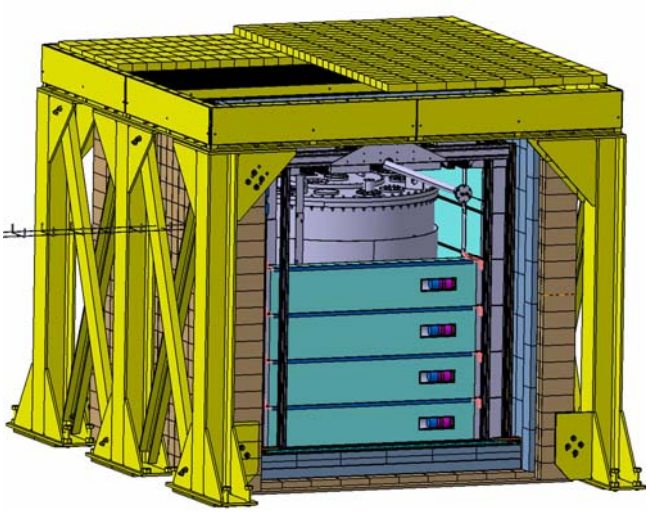

Figure 1. Design of the NUCIFER detector contained in the shielding and muon-veto.

Figure 2. Left: design of neutrino target detector. Right: interior of one module of the muon veto

The overall footprint, including shielding and veto detector, is about 2.8 × 2.8 m^2. Such a compact target volume is very simple, safe and will ensure high detection efficiency. The neutrino detection

efficiency is limited by the detection efficiency of the prompt e+ and of the delayed radiative capture on Gd of neutrons generated by the inverse β-decay reaction. From the simulated detector response, this efficiency results ~50% when we apply a threshold of 2 MeV on the e+ detected energy and of 4 MeV on the gamma cascade detected energy.

3.1. Road-map

The NUCIFER detector with its neutrino target, muon veto and shielding are ready to be installed close to the Osiris research reactor at the CEA of Saclay. Osiris is a 70 MWth reactor. We will install the detector at 7 m from the core at a depth of 5 meter water equivalent (m.w.e.). The data taking will start end 2011 for about 1 year to test the NUCIFER response to reactor neutrino. Regular reactor OFF periods (10 days after each 20 days cycle) will allow studying the spectrum of cosmic background and the efficiency of its rejection.

A further step will be to deploy NUCIFER close to a commercial nuclear reactor, possibly in a country under Safeguards to finally demonstrate the potential of the NUCIFER concept, possibly in collaboration with the IAEA.

4. Nucifer performance

4.1. Neutrino target calibration

For tests, the neutrino target detector has been filled by unloaded liquid scintillator composed of linear alkyl benzene (LAB) with PPO (2 g/l) and bis-MSB (20 mg/l) at the end of 2009. In 2010 its performance have been studied and the calibration performed by using some sources (Americium-Berylium, Cesium 137, Cobalt 60 and Sodium 22) inserted in the centre of the active volume by the Teflon coated stainless steel calibration tube. The measured and simulated calibration curves are displayed in figure 3 and show a excellent agreement, within a few percent between 667 keV and 5.5 MeV.

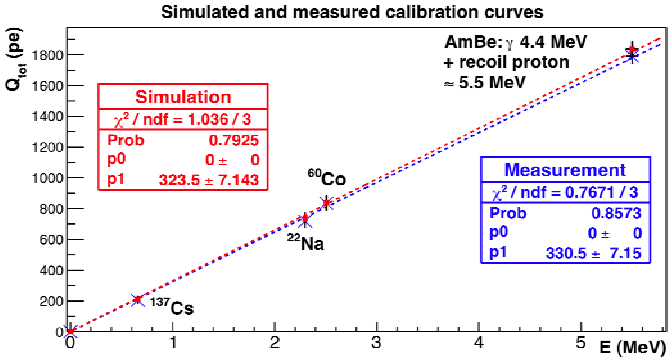

Figure 3: Results of the calibration of the energy reconstruction of gamma ray and neutron events in the unshielded NUCIFER detector (blue) compared with the simulation (red).

By using an Am-Be radioactive source emitting neutrons in coincidence with 4.4 MeV gammas, with a rate of 90 Bq, we were able to mimic the neutrino signal consisting in two correlated pulses within few 100μs. Figure 4, left, shows the distribution of the delay time between the prompt 4.4 MeV gamma and the delayed neutron candidates. Experimental results have been fitted with a sum of an exponential and a constant, the first with a 209 μs time constant attesting clearly the neutron capture on hydrogen, and the second being consistent with the expected accidental background of our detector (naked vessel without shielding). Figure 4, right, shows the energy spectrum measured (black curve) from this source compared with the simulated one (red curve). The right bump shows the sum of prompt energies deposited by the gammas and the neutron induced recoiling protons. The peak on the left is

the 2.2 MeV gamma emitted by neutron capture on Hydrogen. Both prompt and delayed signals, which are similar to the ones expected in case of neutrino detection, are well reproduced by the simulation.

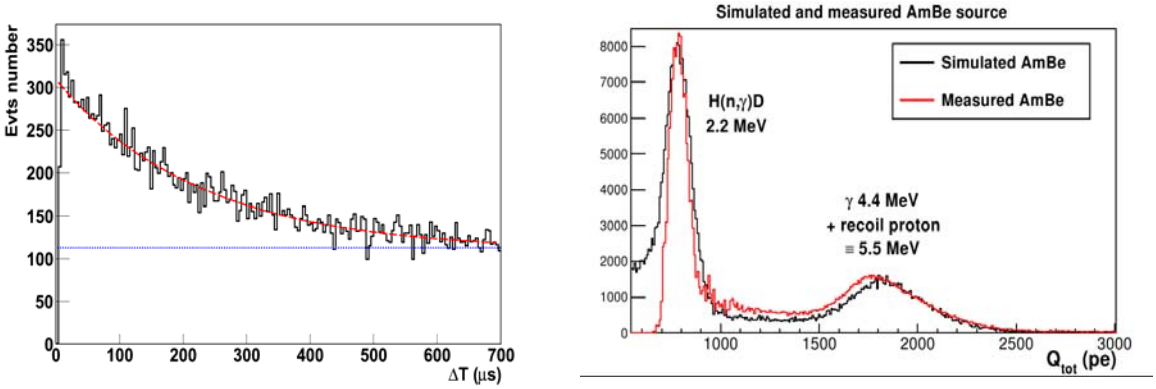

Figure 4: Results of neutron calibration of the unshielded NUCIFER detector using an Am-Be radioactive source. Left : the plot displays the time delay between the prompt 4.4 MeV gamma ray and candidates of a neutron capture. A clear exponential decay with a 209 μs time constant (red dashed curve) appears on top of the flat uncorrelated background (blue dotted line), attesting for the neutron capture signal in our unloaded liquid scintillator (LAB). Right :Measured energy spectrum (black curve) is compared the simulated one (red curve)

4.2. Pulse shape discrimination (PSD)

The main source of background in Nucifer is the correlated one caused by fast neutrons induced by cosmic ray muons. We studied the possibility of using the pulse shape discrimination to disentangle the neutrino signal from these neutrons, in the case they are not tagged by the veto detector. This method is based on the fact that a proton recoil (prompt signal of correlated background) in the final Gd doped liquid scintillator of Nucifer (Eljen-EJ335-05) gives origin to a signal with a shape different from the one induced by a positron (prompt of neutrino signal). This is due to the longer decay time of scintillator for particles with higher dE/dx which translate into a longer tail in the final signal.

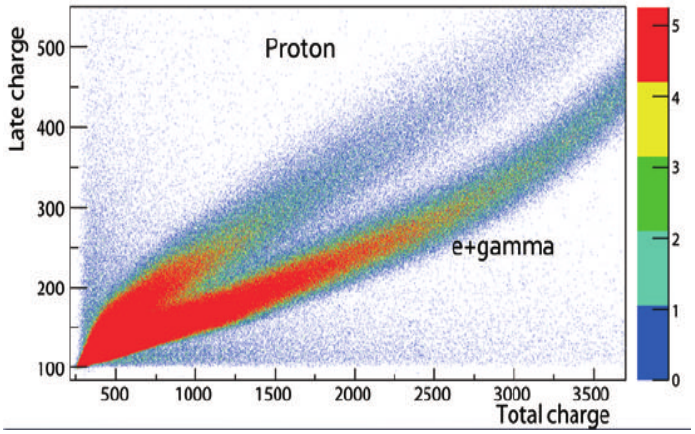

Figure 5: Result of PSD test on the Gd doped liquid scintillator of Nucifer: plot of the charge contained in the tail of the signal as a function of its total charge. We can clearly identify the two different zones for proton recoil and positron signals. Expected detection threshold is around 1500 on horizontal axis.

The PSD capability of the Nucifer liquid scintillator as measured on a dedicated bench (100 cm^3 liquid cells) is shown in figure 5: by plotting the charge contained in the tail of the signal as a function of its total charge we can clearly identify two different zones for proton recoil and positron signals.

The final performance of the PSD will be affected by the volume effect of the Nucifer target. Complementary measurements using the Am-Be source are in progress to quantify the relative loss.

4.3. Remote control

Among the IAEA specifications there is the request of a remote controlled, simply to be used detector. We worked on a remote control of acquisition system, PMT high voltage setting and monitoring, run start and slow control. The data output and possible alarms can be also monitored on-line by a simple understanding display.
The detector stability and linearity are monitored at the 1% level using 4 independent LEDs.

7. Conclusions

The NUCIFER neutrino experiment aims at demonstrating the possibility of high accuracy, reliable, and temper-proof monitoring of fission nuclear reactor thermal power and detecting undeclared Plutonium retrieval. The detector has been tested in an almost final configuration and calibration preliminary results indicate a good understanding of the detector time and energy responses. This attests for the readiness of NUCIFER for the reactor antineutrino hunt. The detector will be integrated at the OSIRIS research reactor at CEA-Saclay during the next fall and will collect first data for the end of the year.

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Nuclear Reactor Simulations for Unveiling Diversion Scenarios: capabilities of the antineutrino probe applied to actual and Generation-IV reactor monitoring.

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Abstract:

In the late seventies was born the idea that nuclear reactor antineutrinos could be used for reactor monitoring. Several experiments in the eighties and nineties, at first devoted to the study of the fundamental properties of neutrinos, evidenced the direct relationship between the detected antineutrino signal and the thermal power and the burnup of the nuclear fuel. The field of applied neutrino physics has shown new developments in the last decade. The International Atomic Energy Agency (IAEA) has expressed its interest in the potentialities of antineutrino detection as a new safeguard tool. Several projects are in development worldwide, designing new antineutrino detector concepts adapted to the non-proliferation constraints. In parallel, sophisticated simulations of reactors and their associated antineutrino flux have been developed to predict the antineutrino signature of the fuel burnup and of a diversion. This prospective simulation work is complementary to the R&D of detection techniques. Considering the actual and future nuclear energy expansion in the world in the next decades, and the various types of future nuclear reactors under design, the antineutrino detection could be among the new technologies that will help IAEA to meet its non proliferation goal. In order to determine how the antineutrino probe could be part of the future surveillance procedures, the characteristics of the antineutrino emission of all these nuclear reactor designs have to be assessed. They will serve as well to determine the sensitivity goal of future antineutrino detectors devoted to reactor monitoring. The IAEA expressed its interest in the study of the performances of the antineutrino technique for safeguarding actual and especially future reactors, with emphasy on Pebble Bed Reactors (PBR) mainly because of the design of their fuel (hundreds of thousands of pebbles in-core) and their online-refueling mode. To this aim we have started to study PBRs with our simulation tools, We use a package called MCNP Utility for Reactor Evolution (MURE), initially developed by CNRS/IN2P3 labs to study Generation IV reactors. The MURE package has been coupled to fission product beta decay nuclear databases for studying reactor antineutrino emission. In these proceedings we will present the state of the art of reactor simulations performed to characterize the antineutrino emission from actual and future reactors.

Keywords: reactor antineutrinos; non proliferation; reactor simulations; Monte Carlo

1. Introduction

Considering the growth of the world energetic demand and the necessity of a reduction of greenhouse gas emission, the world energetic production should grow very significantly in the next decades. Even with an unprecedented development of renewable energies, it is anticipated that the nuclear energy could represent up to 25% of the total energy in 2050. Innovative reactors have to be designed, with

new criteria to meet in order to improve the acceptability of nuclear energy for the next generations, as the ones defined by the Generation IV initiative [1]: improvements on nuclear waste management, fuel economy, economy, non proliferation, safety, sustainability. The IAEA is the United Nations agency in charge of the development of nuclear technologies worldwide, and also of the verification of their peaceful use. Checking the absence of misuse of nuclear facilities is the task of the IAEA's safeguards department. The 11th symposium of this unit which took place in Vienna in November 2010, entitled "Preparing for Future Verification Challenges" [2] has highlighted the main issues the IAEA will face in near future. Indeed, the safeguards inspectors anticipate a spread of nuclear power plants to be inspected worldwide much faster than their financial support. To cope with this increasingly challenging situation, two main concepts are put forward: "remote safeguard inspection" (RSI) and "safeguards by design" (SBD) in order to achieve the "*deterrence of the proliferation of nuclear weapons, by detecting early the misuse of nuclear material or technology, and by providing credible assurances that States are honoring their safeguards obligations*". Thus, an ideal future safeguard tool would be remote, unattended, included during the construction of a nuclear facility, and last but not least, accurate enough to detect a diversion of a Significant Quantity (SQ – cf table 1) of nuclear material in a timely fashion. The timeliness depends on the nature of the material composition and form and on whether it has been irradiated or not.

Nuclear Material	Significant Quantity	Timeliness of Detection
^{235}U from LEU uranium (<20%)	75kg of ^{235}U	1 year
Pu (^{238}Pu <80%)	8kg	3 months
^{233}U	8kg	3 months

Table 1: Significant Quantities of fissile material and timeliness detection (all materials irradiated in-core – source IAEA safeguards glossary 2001))

During the fundamental quest started more than 50 years ago in order to understand the neutrino properties, was born the idea that antineutrinos produced at reactors could carry a direct image of the core, outside the reactor, that could be exploited for a remote control of nuclear power plants [3]. Indeed, large quantities of antineutrinos are produced in the reactor due to β decays of the fission products and about $10^{21}\,\bar{\nu}_e$ /s are emitted in a 1 GW_e reactor core. The distribution of fission fragments depends on the fissile isotopes (^{235}U, ^{238}U, ^{239}Pu and ^{241}Pu) and on the energy of neutron flux in the core. The released energy per fission, the average number of emitted antineutrinos and their mean energy depend also directly on the fissile isotope that undergoes fission, cf. Table 2. Consequently, all the differences in the fissioning tread lead to significant discrepancies in the associated antineutrino spectrum which will reflect the thermal power emitted by the core and its composition.

	^{235}U	^{238}U	^{239}Pu	^{241}Pu
Released energy per fission (MeV)	201.7	205.0	210.0	212.4
Mean energy of antineutrinos (MeV)	1.46	1.56	1.32	1.44
Number of antineutrinos per fission (E>1.8 MeV)	5.58 (1.92)	6.69 (2.38)	5.09 (1.45)	5.89 (1.83)

Table 2: Main characteristics of antineutrinos originating from ^{235}U, ^{238}U, ^{239}Pu and ^{241}Pu fission, in standard PWR [4].

Two pioneering experiments performed at the Rovno power plant in the former USSR and at the Bugey power plant in France have demonstrated the proportionality between the antineutrino counting rate and the thermal power measured by the operators [3,5]. Mikaelian et al. have demonstrated the direct relation between the antineutrino flux and energy spectrum with thermal power and fuel content of a reactor core [6]. At a fixed power, the neutrino flux as well as the shape of the energy spectrum are affected by any change of the fuel composition. As an example, a hypothetical reactor able to use only ^{235}U would produce an antineutrino flux 40% higher than the same reactor burning only ^{239}Pu.

Linked with the energy dependency of the detection interaction cross-section, the detected neutrino flux would be 60% higher. Estimation of the thermal power hence requires the knowledge of the fuel history (initial composition) and the simulation of its evolution in time.
Antineutrinos not only provide informations on the reactor power but also on its isotopic content, opening several application possibilities such as burnup monitoring for fuel economy and safeguards aspects.
A worldwide effort concentrated on applied antineutrino physics has started a few years ago, in the US, France, Brazil, Japan, Russia, Corea, and Italy [7] to further investigate the feasibility of the use of such detectors for safeguards purpose. Recently, with a cubic meter sized antineutrino detector using Gd-doped liquid scintillator as a target, it was demonstrated that it is possible to monitor precisely the thermal power over hour to month-time scales in a non intrusive and unattended mode. In addition, it was proved that antineutrino detection enables to get information on the plutonium content of the reactor core [8].
In October 2008, the New Technology department of IAEA organized an international expert meeting on Antineutrino Detection for Safeguards applications. Discussions between physicists from all the quoted above collaborations and the inspectors of the IAEA were carried out, defining a list of potential applications of antineutrino detectors at reactors if this novel technology is approved [9]. The reactors and cases which should be addressed were listed as: on-load reactors such as CANDU and VHTR (PBMR) ones, PWR and BWR, research reactors, innovative fuel use, future reactors (Generation IV or other concepts). Among the types of reactors chosen by the Generation IV forum, the Pebble Bed (PB) reactors have been identified as challenging for non-proliferation issues mainly because of the design of their fuel (hundreds of thousands of pebbles in-core) and their online-refueling mode. Thus, antineutrino measurements could comprise a unique means of providing information on the isotopic composition of the core, non-intrusively and in near real-time.
In 2008 were also discussed the IAEA requirements for a reactor monitor tool based on neutrino detection. Such a detector should be relatively small, portable, cheap, safe and remote controlled. These detection requirements constrain also the sensitivity of the antineutrino probe. Thus, to infer to which extent antineutrinos could provide a diversion signature, the characterization of the antineutrino source associated to different contemporary or future reactor designs and fuels is mandatory. This is to be the first step of our feasibility study and necessitates the use of simulation tools.

In order to define the contribution of an antineutrino detector to the existing and future safeguarding procedures, we have undertaken a few years ago the simulation of different reactor concepts taking into account reactor physics that constrains strongly the possible diversion scenarios. We will present here as a review the different reactor designs for which we have studied the associated antineutrino emission and computed diversion scenarios, as well as the ones for which this work is on-going.
To this purpose we use the MURE code interfaced on MCNPX that we have coupled to the existing nuclear databases as well as to recent experimental results concerning fission product beta decay properties in order to compute the beta and antineutrino emission through a summation method.
In these proceedings, the antineutrino emissions from a PWR, a CANDU reactor and a Generation IV concept : a Pebble Bed Reactor (one of the 2 main Very High Temperature Reactor designs studied in the world) will be presented successively, after a short presentation of our simulation tools.

2. Simulation method and tools

To characterize the antineutrino source associated to a reactor, the evolution in time of the concentration of the fissile nuclei and of the fission products in the core is needed.
For that purpose, we use the MCNP Utility Reactor Evolution (MURE) package. Designed by CNRS/IN2P3 laboratories for the simulation of Generation IV reactors, MURE [10] is a precision, open-source code written in C++ which automates the preparation and computation of successive MCNP (Monte-Carlo N-Particle) calculations and solves the Bateman equations in between, for burnup or thermal-hydraulics purpose. Because of these abilities this tool is also suitable for our study of the flux and energy spectrum of the neutrinos emitted from various core designs. MURE is available at NEA (NEA-1845). The evolution in time of the isotopic composition of the core is driven by the initial fuel composition, the input of the thermal power and the refueling scheme. It is perfectly adapted to take into account the physics of a reactor core, especially the neutronics as neutron capture in various nuclei in the core give non-negligible distortion of the total energy spectrum [10]. For our specific purpose, we have adapted the code to compute and to store the amount of all β- emitters produced over time. The simulated antineutrino energy spectrum and flux obtained were first compared with

pure U and Pu isotope target experiments for validation [11]. Some comparisons of the nuclear evaluated databases have been performed and new nuclear experiments aiming at measuring beta decay properties of fission products have been proposed [12].

3. Antineutrino emission from PWR, CANDU and Pebble Bed Reactors

3.1. PWR antineutrino emission

Being the most standard reactor type used in the world, a full core simulation of a PWR reactor has been performed to compute the fission product concentrations. In a PWR, the initial fuel usually consists in enriched uranium, with three different ^{235}U enrichments : 1.8, 2.4 and 3.1% placed in the core so as to flatten the neutron flux. Fuel rods are coated with 0.6 mm Zyrcaloy cladding, which are properly taken into account in the simulation. Each assembly constitutes a square lattice of 289 cells (17x17) of dimensions 12.6x12.6x4796 cm3. In each assembly, 25 guide tubes are present for instrumentation (central rod) or absorbant or burnable poison rods. At the moment, the simulation of the control rods has not been performed as it would induce huge CPU times. Then, in our simulation, the criticality is only controlled with Boron diluted in the moderator, which is about 1000 ppm and constant over time. The water and cladding temperature is 600 K, and the effective fuel temperature considered is 900 K.
The inventory is then obtained for a standard N4-PWR loaded with fresh fuel and without refueling. The power is maintained constant to 4.27 GWth,. As soon as the reactor is operating, reactions of neutron capture on ^{238}U produce ^{239}Pu and ^{241}Pu, which then also contribute to the energy production. The main contribution comes from the two isotopes ^{235}U and ^{239}Pu, which contribute to 90 % of the fission rates. Since the neutron flux is mostly thermal, and due to the very low relative evolution of ^{238}U present in the core, ^{238}U makes a small and constant contribution. The ^{241}Pu isotope gives also a small contribution to the total power, increasing with time as ^{241}Pu is produced through neutron capture.

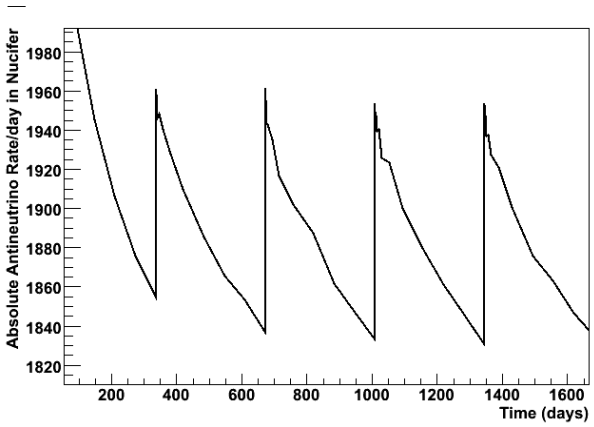

Figure 1: Antineutrino rate per day detected in a 50% efficiency liquid scintillator detector (target 1.1t, without energy threshold)) placed at 25m from the core.

During the refuelling of such a PWR, 900kg of ^{235}U is added in the core while 250kg of ^{239}Pu is retrieved, resulting to a variation of 6.5% in the detected antineutrino flux for an equilibrated core (after 2 refuellings), as shown in Fig. 1. In Fig. 1 a concept of antineutrino detector based on a Gd-doped liquid scintillator 1.1t target placed at 25m of the reactor core is assumed. A detection efficiency of 50% is assumed as this is the efficiency goal for the detector Nucifer of very similar design [13]. The considered distance of 25m between the detector and the reactor core is the one adopted in the SONGS experiment [8].

3.2. CANDU antineutrino emission

CANDU reactors use heavy water as moderator and coolant and natural Uranium fuel. Moreover they are on-load reactors. For these two latter features, they are considered as potentially proliferant reactors. This is the reason why we started a simulation of a channel of a CANDU 600 reactor. Such a channel is called a force tube, there are 380 such pieces in a CANDU 600 reactor, each containing

12 fuel bundles. The simulation of a fuel bundle was already simulated with the MURE code and compared successfully with a simulation using the deterministic code DRAGON in A. Nuttin et al. [14]. We have performed a similar simulation of a fuel bundle and compared it to the simulation of A. Nuttin (LPSC Grenoble), obtaining similar results.
The dwell time is defined by the time when the k_∞ decreases under the value k_∞=1, taking into account 5000 pcm for neutron leaks and absorptions. The influence of the moderator, coolant and fuel temperatures are taken into account. The shown results are obtained with $T_{coolant}$= 600 K, $T_{moderator}$ = 300 K and T_{fuel} = 1200 K. The obtained dwell time of a bundle amounts to about 200 days, when taking into account the neutron leaks (3000 pcm) and the absorptions in the core (by Boron for instance, 2000 pcm), which are not accounted for in this simulation. These values have been carefully estimated by [14] thanks to a full core simulation.
The bundle simulation (Fig. 2) takes also into account 3 different radial zones for the 37 pins, to reproduce more accurately the neutron flux distribution, as was stated in A. Nuttin et al. [14].

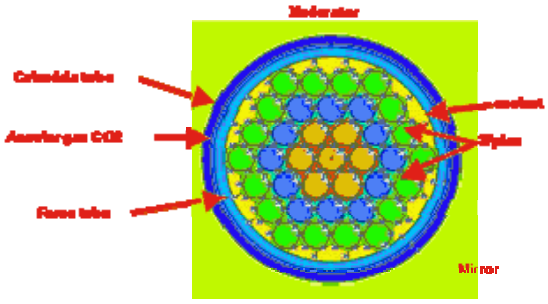

Figure 2: Simulation of a fuel bundle. The mirror is placed so as to reproduce the moderation ratio of a CANDU 600 reactor.

Giving these results, a fuel channel has been simulated, approximating 4 bundles by a “giant bundle” of the same dimensions, neglecting then the cladding in between the concerned bundles, which should be a good approximation as this material has quasi no influence on the neutronics. The total amount of fuel put in the simulation however corresponds to the one of a CANDU 600 channel. The infinite multiplication coefficient factor, the neutron flux distribution and the obtained inventory have been compared with a similar simulation performed independently by A. Nuttin and still good agreement was found between the two simulations. The details of the neutronics associated to this simulation can be found in [15].

Given these results, several channels of a Heavy Water Reactor (CANDU 600), loaded with natural Uranium and with various fuel dwell-times have been simulated also in order to provide a first hint of what antineutrino detection would bring to the monitoring of such on-line refueled reactor which are maintained in a steady state through quasi-continuous refueling. Very simple proliferation scenario studies could then be computed in a second part to show the abilities of the tools we have developed and give a first hint of the capabilities of the antineutrino probe to monitor such reactors.
The philosophy we adopted is to divert Pu of quasi military quality by refuelling more often some channels, and in the mean time refuelling slower the same amount of channels to mask the diversion, conserving the total number of daily refuelled channels in the reactor. The time between two consecutive refuellings of the two thirds of a channel (so-called "channel refuelling time" in the following) is about 200 days, and as a mean 2 channels are refuelled per day in the reactor. The isotopic vector of Plutonium for a residency time of the fuel of about 100 days is close to military quality. Therefore, for sake of simplicity, we chose to refuel each channel after 100 days and 300 days. This case has to be compared with a refuelling scheme of all channels every 200 days. In each case, two channels per day are refuelled by two third. We paid attention to the fact that the total reactivity associated to both is conserved. This comes from the nearly linear dependence of the k_∞ after 50 days [15].
As stated above, this scenario is voluntarily unrealistic in order to test the antineutrino detector response to a large diversion case, and meanwhile demonstrates that our simulation tools are able to simulate such a scenario. The refuelling of a channel concerns usually two third of a channel, 8 bundles are sent to the pool, while the remaining irradiated 4 bundles are pushed toward the extremity of the channel, and 8 fresh fuel bundles are inserted. The MURE code allows to simulate such a refuelling scheme for different periods. The obtained inventories for a 200 days refuelling scheme of a channel as a function of time are shown in Fig. 3. The large variations happening periodically

correspond to the refuelling of the channel, a periodic inventory is obtained after the first channel refuelling. Fig. 4 shows the resulting antineutrino interaction rate through the inverse beta decay process, in a hypothetic detector filled in with 1.1t of Gd-doped liquid scintillator.

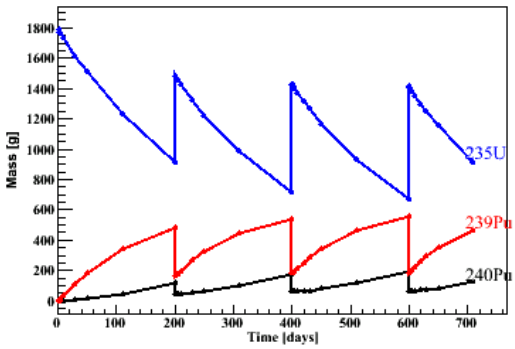

Figure 3: Inventory of one channel simulated with MURE, with 100, 200 and 300 days channel refuelling times.

In order to simulate the normal reactor refuelling scenario, the results of the time dependence of the antineutrino flux from 400 channels (instead of 380 for simplicity sake) have been added, with their history in the core shifted by one day, and refuelling 2 channels a day. The adequacy between the number of channels, the 200 days channel refuelling time and the daily refuelled number of channels implies a steady state of the reactor, resulting in a flat daily antineutrino rate as a function of time in an antineutrino detector. The number of antineutrino interactions in the target through the inverse beta decay process amounts to 2236 in a Gd-doped liquid scintillator detector assuming a target of mass 1.1t. The detection efficiency has to be taken into account to obtain a realistic antineutrino detection rate. A detailed study of the associated simulation errors has been undertaken and is on-going. The statistical error due to the use of a stochastic code is of the order of 1%.

The same exercise has been performed in the diversion scenario case, with 200 channels refuelled every 100 days and 200 channels refuelled every 300 days, keeping the 2 refuelled channels per day frequency. A steady state of the reactor is obtained also, but amounting to a lower antineutrino rate, in detector of design such as the one described above, of 2157 antineutrino interactions per day (with the same error as quoted above), as the respective number of fissions in both reactor operations are different.

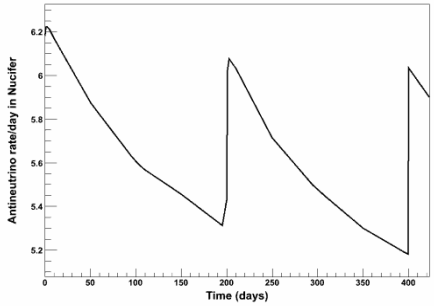

Figure 4: Fitted antineutrino rate per day in Nucifer placed at 25m from the core, corresponding to the Fig. 3 channel inventories as a function of time, for 200 days channel refuelling times.

The discrepancy between the two rates amounts to a relative difference in the antineutrino rate in Nucifer of 3.5%, provided that a detector filled with 1.1t of Gd-doped liquid scintillator can reach a 1% level statistic accuracy within approximately 10 days and that relative measurements would allow to cancel most of the systematic errors. In the diversion scenario, 110 kg of ^{239}Pu were retrieved from the channels with refuelling time 100 days. The retrieved plutonium vector contains also 20.4 kg of ^{240}Pu and 40g of ^{238}Pu. These results have been presented at the IAEA Symposium on International Safeguards: Preparing for Future Verification Challenges, in November [16]. As the retrieved plutonium is not weapon-grade, we have thus performed another diversion scenario with refuelling schemes at 50 days and 350 days. In this scenario the discrepancy in the antineutrino rates with the

normal refuelling scheme should moreover be enhanced. The study of this diversion scenario is on-going.
Of course, CANDU reactors are operated in adapting the channel refuelling time of the channels to flatten the neutron flux in the core. Control rods, and light water are used also for this purpose. This study only shows that for a steady state reactor, operated at constant power, such as CANDU ones, a large difference in the refuelling frequency would lead to a different rate in an antineutrino detector because of the different burnups of the channels compared with a normal operation of the reactor. We can deduce also from these results the order of magnitude of the Nucifer sensitivity to a ^{239}Pu diversion, keeping in mind that the Nucifer detector will take data in fall 2011 and that the systematic errors associated to the detector will be carefully assessed from this time.

3.3. Pebble Bed reactor antineutrino emission

The study of the ability of the antineutrino probe close to actual nuclear power and research plants is mandatory in order to develop adapted detector designs and test them experimentally. Their performance could then be compared with existing and efficient-proved systems used by IAEA nowadays. As regards future reactor designs, a new safeguards policy is under development based on Safeguard by Design and Remote Safeguard Inspection methods, among others. The antineutrino detection technique could meet the requirements of these two methods, given the nature of these particles and the characteristics of the detectors under development. It is thus also mandatory in order to chart a path toward the inclusion of the antineutrino probe into the set of safeguards tools used by the IAEA to study the characteristics of the antineutrino emission from the main Generation IV reactor designs and deduce what could be the specific contributions of this technique to the IAEA safeguarding system.

Among generation IV prospective nuclear plants, Very High Temperature Reactors (VHTR) present the interest of being multi-skilled. They operate at very high temperatures, improve the efficiency of the reactor for electricity supply, but allow also the production of Hydrogen or the replacement of petroleum fuel in industrial high temperature processes. This concept has been of interest since early in the 40's: in the UK (Dragon), USA (Peach Bottom, Fort Saint Vrain), Germany (AVR, THTR-300) and Japan (HTTR). South Africa and China have also developed studies for such reactors. (PBMR in South Africa and HTR-10, HTR-PM in China). Chinese research reactor HTR-10 has performed its first criticality in 2003. There are two concepts of VHTRs under study worldwide: the prismatic or the pebble-bed ones. Both variants use small coated particles (CP) of fuel, the coat being made of SiC: in the latter, these CPs are packed in graphite matrices in the form of pebbles of the size of a tennis ball. In PBRs, the pebbles are put in-core from the top of the reactor and go down the core by gravity, a given number of pebbles being withdrawn every day. At their withdrawal, the pebbles undergo a test on their burnup, and if the target burnup is not reached, the pebble returns to the core for another pass in-core [17]. Thus, each pebble undergoes several passes in the core before it reaches its target burnup, the number of passes depending on the optimization calculations specific to each core. The target burnup of a pebble reached in VHTR is higher than in the fuel of nowadays PWR, which is another advantage of the VHTR concept. The study of neutrino detection applied to this kind of reactor is of interest for the IAEA as it concerns an on-load refuelling concept. Following the initial fuel loading, the PB reactor will reach an equilibrium condition that should persist for the operating life of the reactor. Operating parameters should then be highly predictable when the reactor is operated under optimal power production conditions. Departures from these conditions may be viewed as potential safeguard significant events. Antineutrino measurements provide information about the isotopic composition of an entire fissioning reactor core – in this sense they provide a type of bulk accountancy for that core. If a Bulk Accountancy safeguards system is adopted for such PB reactors, antineutrino measurements could provide a unique means of providing such measurements non-intrusively and in near real-time. Following the emphasis put on this specific concept at the meeting devoted to the antineutrino detection, hosted in Vienna at the IAEA headquarters in 2008, a simulation work has been started with the aim to study diversion scenarios at PB reactors with antineutrino detection.

As a first step for our computations, we have compared our simulation to a benchmark organized by the Nuclear Energy Agency (NEA) [18]. We have simulated a single cell of a pebble containing various types of fuel: UOx, PuOx and ThUOx.

The main difficulty in simulating this kind of reactor is the treatment of the so-called "double heterogeneity" due to the random distribution of the CPs in the pebble and the random distribution of

the pebbles in the core. While MCNP5 computes random geometries, MCNPX does not. An estimation of the errors due to the approximation of a random geometry with a regular lattice was published by Bomboni *et al.* [19] and concluded that the errors are negligible as long as the initial error in the fuel inventory due to the difference in the geometries does not exceed 1%.

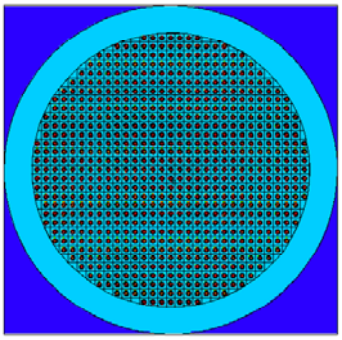

Figure 5: Geometry of a single pebble surrounded by Helium in a cubic cell

In the benchmark, each pebble contains 15 000 CPs of fuel (with a radius of 0.025cm for UOx, and 0.012cm for PuOx and ThOx) in a fuel zone of graphite of 2.5 cm of radius (cf Fig. 5). Our use of a regular lattice leads to 14939 CPs in a pebble, which corresponds to an error in the initial inventory of less than 1%. Each pebble provides 556.25W and there are 359548 pebbles in the reactor core described in the NEA benchmark leading to a 200MWth reactor power. The UOx fuel is enriched at 8.2%, and there is 10.21g of UOx (9g of heavy nuclei) in the pebble. Our simulation, with a1000K fuel temperature using the JEFF2.2 nuclear databasis, compared with WIMS9/JEFF2.2 leads to discrepancies in inventories of less than 6% for ^{235}U and ^{239}Pu, and less than 10% for ^{241}Pu. More detailed results about our PBR simulation can be found in [20].

From the inventories of the fissile nuclei in core and their fission rates, we can compute the evolution in time of the flux of antineutrinos emitted (cf Fig. 6).

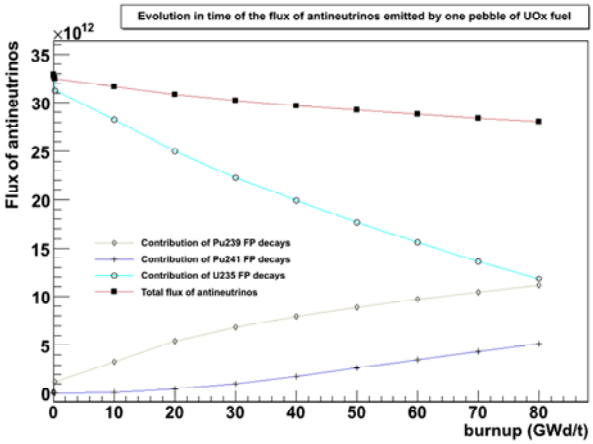

Figure 6: Evolution of the flux of antineutrinos as a function of burnup for UOx fuel (in red, total flux, in sky blue ^{235}U contribution, grey, ^{239}Pu and navy blue ^{241}Pu).

In this section, the fluxes of antineutrinos presented are the multiplication of the reaction rate of each fissile nucleus by the mean number of antineutrinos with an energy above 1.8MeV emitted after its fission (from the data in table 1). Considering the case of ^{233}U, we have taken the data of ^{239}Pu as Hayes *et al.* [21] found both nuclei had very similar antineutrino emission characteristics using the fission product decay database ENDFBVI.8. This first approach will shortly be refined thanks to the MURE package, by building the antineutrino flux from the fission products inventories and their beta decay data. The procedure we have developed for that purpose is described in ref. [15].

As an indication of the influence of the uncertainty associated to the simulated fission rates on the deduced antineutrino flux, we have compared the latter flux built from the inventories obtained with

WIMS9/JEFF2.2 with the one obtained with MURE/JEFF2.2. The obtained fluxes differ by less than 0.5%.

We have applied the same procedure to PuOx fuel and ThUOx fuel. In these cases, there is 1.129g of reactor grade plutonium (resp.ThUOx). Our simulations are in good agreement with the benchmark, with discrepancies in the main fissile nuclei of less than 1% compared to the participants. In Fig. 7, we present the difference of antineutrino flux between PuOx pebble or ThUOx pebble compared to UOx pebble.

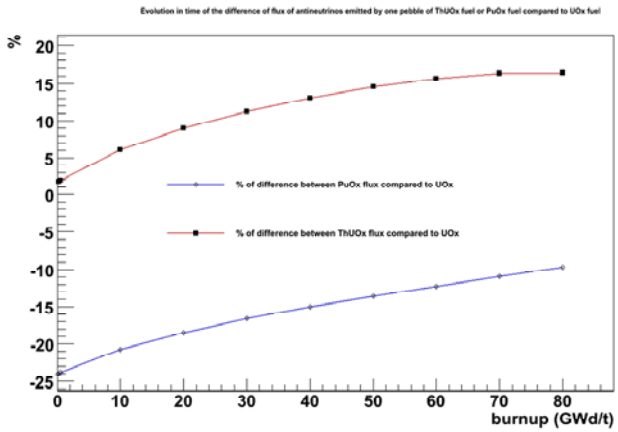

Figure 7: Evolution in time of the difference in antineutrino flux emitted by a pebble of PuOx (in blue) and ThUOx (in red) compared to UOx.

3.3.1. Diversion scenario for UOx fuel

From the single mirrored pebble of the benchmark, representing an infinite reactor, we need to get a first estimate of what would be the flux emitted by a real reactor, and, in this first scenario, we will focus on the flux at steady state. To this purpose we need to compute

At first, we have evaluated the neutron leakage from the pebbles in order to evaluate the dwell time of each pebble in-core. We have thus developed a simplified full core simulation in order to compute the value of the effective multiplication coefficient k_{eff} associated to the packing fraction of the pebbles in the considered core (61%). The discrepancy between the obtained k_{eff} and the infinite k obtained with the pebble simulation gives a neutron leakage amounting to 11%. We have considered in addition an antireactivity of 4500 pcm to account for the control rods. From these calculations, we could deduce a dwell-time of the pebbles of 80GWd/t, which leads to an average burnup in-core at equilibrium of 40GWd/t, in agreement with the values found in literature. The average burnup will allow us to compute the emitted antineutrino flux associated to the normal operation of the reactor core.

As these approximations need refining because of the complex pebble management in these reactors, we have computed our first scenarii for a core at equilibrium at 40GWd/t, and also for 50 GWd/t, in order to check the influence of these hypotheses on the results of the diversion scenarios. We have first estimated the number of pebbles to divert in order to get a SQ of fissile material as a function of burnup, in order to check which fuel takes fewer pebbles to divert: the diversion of PuOx becomes more interesting for a burnup of approximately 20 GWd/t (cf. Fig. 8).

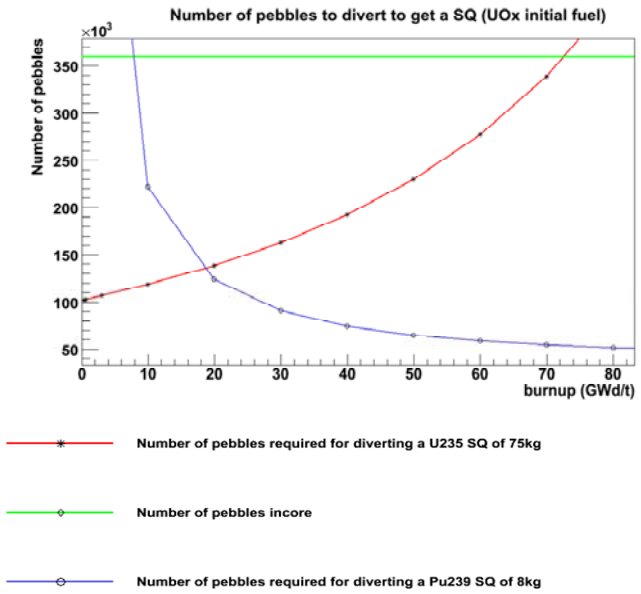

Figure 8: Number of pebbles to divert to get a SQ for each fissile material as a function of burnup (in blue, SQ of plutonium; in red, SQ of ^{235}U. The green line represents the number of pebbles in-core).

From this, we can build the influence of a diversion of a SQ of fissile material on the average flux as a function of burnup, for the average burnup at 40 GWd/t (see Fig. 9) and 50 GWd/t.

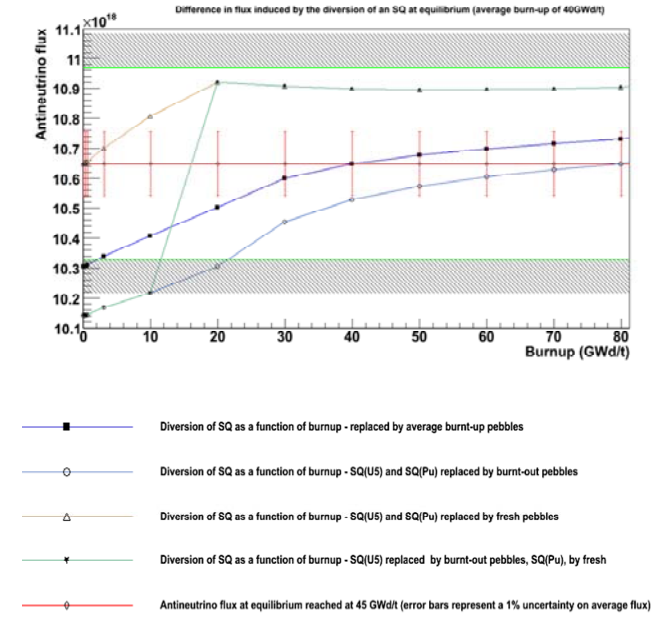

Figure 9: Influence in flux of a diversion of a SQ for an average burnup of 40GWd/t

The dashed areas materialize discrepancies of more than 3% from the flux reached at equilibrium. The dark green line corresponds to the replacement of fresh pebbles by burnt-out ones, and the replacement of any pebble containing plutonium by fresh pebbles.

The same cases have been studied for a burnup at 50 GWd/t. It is to be noticed that the main discrepancy found due to the shift of average burnup in the core concerns the steady-state flux itself, which is here of 1.8%, while the influences of diversions remain ranging from 0% to 3.5% as in the case of an average burnup of 40GWd/t.

3.3.2. Diversion scenario for PuOx or ThUox fuels

A first simplistic scenario would be the diversion of fresh PuOx or ThUOx pebbles that would be replaced by fresh UOx pebbles, at the startup of the reactor.
During that phase, half of the pebbles are dummy (graphite) pebbles. As there is about 1g of fissile material in PuOx pebbles, it requires 8000 pebbles to get a SQ, whereas the mass of fissile material in ThUOx is much smaller, implying a diversion of significantly increased number of pebbles (see Fig. 10).

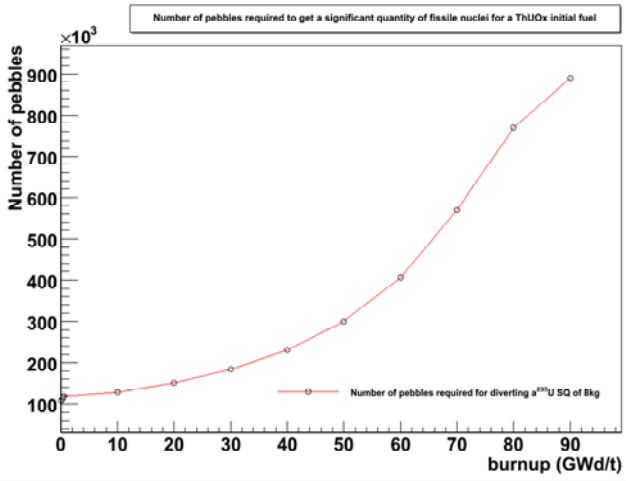

Figure 10: 108000 fresh pebbles contain a SQ of ^{233}U

From the discrepancies evaluated in Fig. 10, one can easily compute that a diversion of a SQ of fresh PuOx pebbles replaced by fresh UOx ones would lead to a discrepancy of 1% to the expected flux of the benchmark reactor. Assuming now that a detector would be sensitive to a discrepancy of 3% from any expected flux, would lead to impose that there are no more than 64 000 fresh pebbles at the start of the reactor, which leads to a maximum power of 72MWth for this reactor.

The case is slightly improved for ThUOx fuel where the maximum power acceptable from the neutrino detection point of view would be of 80MWth.

3.3.3. Application to the study of a PBMR

Ref. [22] is a safeguard study from INL lab to IAEA's profit about South Africa PBMR taking into account the rescaling of the project from 400MWth to 200MWth: we have simulated a PBMR pebble, which contains 9g of heavy nuclei, of uranium enriched at 9.6%. The target burnup of the pebbles is of 90GWd/t with a steady state of the reactor reached at 45GWd/t.

We have computed the same diversion cases and studied the same figures as Fig. 9 for the 400MWth and the 200MWth versions (see [20] for a more detailed discussion). These preliminary studies prove that another key parameter to take into account is the ratio of the number of pebbles required to get an SQ compared to the number of pebbles in core, which is directly linked to the thermal power of the reactor, its power density, the mass of fuel per pebble and the enrichment of the fuel.

For instance, a Gd-doaped liquid scintillator detector of target mass 1.5t, placed at 25m from the core, would be able to detect, with a careful control of the systematics leading to a total error on the flux measurement of 2.5%, a diversion in a timely fashion (less than 3 months) in the following cases :

- replacement of fresh pebbles by burnt-out ones until 25GWd/t

- replacement of irradiated pebbles by fresh ones from 20GWd/t.

The results obtained with the PBMR of 200MWth and the 200MWth PBR of the NEA benchmark are very similar. The PBMR fuel is a little more enriched, leading to slightly better results with a discrepancy in the emitted antineutrino flux for the diversion of an SQ greater than 3%. The 400MWth version of the PBMR gives a discrepancy in the emitted antineutrino flux for the diversion of an SQ lower than 2%, showing that the absolute sensitivity of the antineutrino probe depends very much on the power level of the reactor.

So far, we have considered the simplistic scenario of a diversion of all the pebbles at one given burn-up, replaced by fresh pebbles (as soon as plutonium is diverted) when the reactor is at equilibrium.
Other data that should be taken into account are the quality of the plutonium as a function of burnup (Fig. 11), in order to determine at which step (which pass of pebble) a pebble would be the best compromise to divert, between the mass of plutonium and its quality. For instance, in the case of the PBMR, the calculation of the inventories show that ^{238}Pu will remain inferior to 2% until the target burnup. These data are to be an input in the calculations of Kessler *et al.* [23]considering the feasibility of setting an explosive device from reactor-grade plutonium (and discussing as well the pertinence of a wild-spread accepted threshold of 6% of ^{238}Pu to consider reactor-grade plutonium proliferation resistant), or process described by King *et al*. [24].

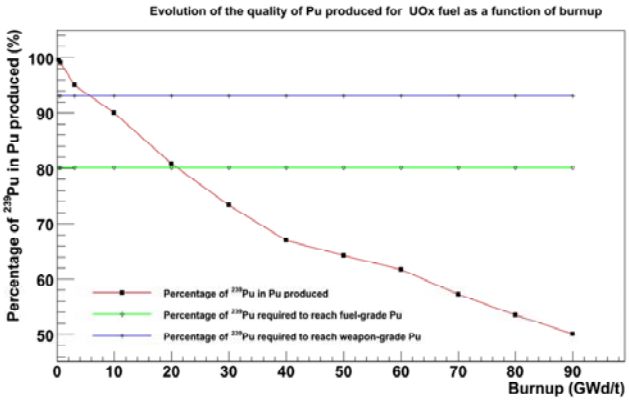

Figure 11: Quality of plutonium as a function of burnup

4. Conclusions and outlooks

We have described in these proceedings the tools we have developed to assess the ability of antineutrinos emitted by reactor cores to monitor nuclear reactors. To this aim, reactor simulations with the MCNP Utility for Reactor Evolution (MURE) code have been performed, and the code has been adapted to compute fission product beta spectra, thus allowing to build the antineutrino energy spectrum and compute the associated flux. Complex reactor simulations, including actual and future reactor designs, are mandatory in order to evaluate the way reactor antineutrino detection could integrate future privileged safeguarding procedures such as safeguards by design or remote inspection safeguards. These simulations and associated diversion scenarios will also bring important indications of the detection efficiency goals that innovative antineutrino detectors under design or construction should reach in order to meet the required deterrence effect.
A generic full-core simulation of a PWR, the most common reactor design in the world, have first been performed in order to compute its antineutrino emission. The simulation of several CANDU reactor channels have been also shown in these proceedings. A gross proliferation scenario has been studied and the response of a Gd-doped liquid scintillator detector has been assessed, showing that such a detector placed at 25m from such a reactor core could detect a variation of about 80kg of the ^{239}Pu inventory. The study of more refined scenarios with CANDU reactors are contemplated. As suggested by the IAEA, the capability of the antineutrino probe for the monitoring of future reactors and the use of innovative fuels deserves a particular attention. We have thus started the neutronics study of a Pebble Bed Reactor (VHTR), starting with a cell calculation, and its associated rough first scenarios of diversion. The refining of the estimation of errors (mainly in the inventories) is under study and their propagation is a key step in order to go further in the study of the antineutrino probe for safeguards issues, as it leads to performances criteria required for the antineutrino detectors to be useful. The study of other Generation IV reactors as well as the OSIRIS reactor close to which the antineutrino detector Nucifer (see A. Porta et al., these

proceedings [13]) is on-going in our team and results should be presented at the next meeting at IAEA devoted to the antineutrino technique development in fall 2011.

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21 Open source and satellite imagery

Open Source Information Acquisition and Analysis in the IAEA Department of Safeguards

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Abstract:

Acquisition and analysis of open source information plays an increasingly important role in the IAEA's safeguards system. Following the approval of the Model Additional Protocol in 1997, the IAEA's focal point for the collection and analysis of open source information is the Division of Safeguards Information Management within the IAEA's Department of Safeguards. Open source information analysis is fully integrated with core safeguards processes and activities, and has become an effective tool in the work of the Department.

Open source collection and analysis is part of the Safeguards Department's evolving State-level concept; a holistic approach to safeguards implementation which ensures that the planning, conduct, and evaluation of safeguards for an individual State are based on an ongoing analysis of relevant information available to the IAEA about the State.

This paper provides an overview of the acquisition and use of open source information in several thematic areas: evaluation of additional protocol declarations; support to the State Evaluation process and infield verification activities, including in-depth investigation of safeguards issues, supporting inspections, design information verification and complementary access; research on illicit nuclear procurement networks and trafficking; and monitoring nuclear developments.

Demands for open source information have steadily grown and will likely continue to grow in the future. Coupled with the enormous growth and accessibility in the volume and sources of information, new challenges are presented, both technical and analytical. This paper discusses actions already taken and outlines future plans for multi-source and multi-disciplinary analytic integration to strengthen confidence in safeguards conclusions, especially regarding the absence of undeclared nuclear material and activities.

Keywords: Open source information; safeguards information analysis; State Evaluation

1. Introduction

Following the rapid advances in the digital world, open source information [1] has become a central component of the working environment in both the private and the public sectors. In the course of progressive development of nuclear safeguards, the IAEA has devoted increasing effort to effective collection of such data, associated evaluation and analysis, as well as proper dissemination of acquired knowledge. The IAEA is in the unique position of being able to combine and compare State-declared information, inspection reports and findings, together with information collected from a wide variety of open sources.

With the approval of the Model Additional Protocol [2] in 1997, comprehensive capabilities for collecting and processing open source information were created in the Division of Information Management (SGIM), a division in the Department of Safeguards which traditionally had been responsible for

processing nuclear material accountancy data and providing hardware and software support for the Department's computer systems. The development of SGIM's capability in this new direction was significantly aided by voluntary assistance from Member State Support Programmes, which, coupled with IAEA core resources, have enabled SGIM to acquire specialized tools, train its staff members and hire individuals with diverse skills and expertise. [3]

2. Open source information and the State Evaluation Process

The Department's open source acquisition requirements are met by continuous monitoring of major open source information providers and by performing specialized collections. In addition, new sources, collection tools and methodologies are regularly evaluated for efficiency and effectiveness and, if found useful and cost-effective, deployed.

Unlike the well-structured nuclear material accounting data provided to the Agency by States in fulfilment of their reporting obligations under safeguards agreements, the collection of unstructured open source information presents several challenges. First, there is an overwhelming and growing quantity of information available on all thematic areas relevant to Agency safeguards. Second, multiple languages and information sources, of widely varying reliability and relevance, must be systematically processed. Third, and more importantly, "raw" information usually requires significant effort by highly trained collectors and experienced analysts collaborating though systematic procedures to review, correlate, evaluate, and prepare synthesized information products and services.

The principal reason for collecting open source information is to support the Department's State evaluation process, instituted in the late 1990s to bolster the credibility of the safeguards conclusions reached by the Agency. This is a regular, (typically annual), in-depth review and evaluation of all safeguards-relevant information available to the Agency, which is conducted to evaluate States' compliance with their safeguards agreements. At present, over 100 information compilations per year are being prepared for the State evaluation process. These so-called "State files" comprise a wide spectrum of information – ranging from background political, economic, legislative regulatory, and security information, to more specialized scientific, technical and other fuel-cycle related data directly relevant to the analysis and evaluation of each State's nuclear programme. In addition, focused collection of information and analytical reports are frequently needed to address safeguards-related issues and questions, evaluate State-submitted additional protocol declarations, and assist with the planning of complementary access and inspection activities and subsequent follow-up. [4] Since the early 2000s, open source information has also been utilized in analysis of transnational nuclear procurement networks. [5]

In addition to contributing to State evaluation and related analytical tasks, open source information is collected for monitoring and "early-warning" purposes. Thousands of information items from a wide variety of sources are filtered through refined search procedures, and hundreds reviewed by analysts on a daily basis, to monitor new and on-going nuclear proliferation and safeguards issues. Selected items of highest immediate relevance are widely disseminated within the Department of Safeguards, via an e-mail newsletter, the SGIM Open Source Daily Highlights. In addition, items of more particular interest are also provided by e-mail directly to relevant staff in the Safeguards Department, according to the specific needs of their work. The daily review also feeds information items into the Open Source Information System (OSIS), the Department's main repository of open source information, which is accessible by all Safeguards staff through a free-text search engine that leverages a nuclear fuel cycle "topic tree" organizational structure.

For States with an additional protocol in force, the Agency has the ability to provide credible assurances about the absence of undeclared nuclear material and activities. Keeping in mind that "absence of evidence" does not necessarily prove the "evidence of absence", the demonstration of such a negative presents considerable information-analytic challenges. State evaluation requires the systematic integration, analysis, and cross-corroboration of various types of information: inspection and complementary access reports; nuclear material accounting reports and additional protocol declarations by States; environmental sampling results; open source information and analysis; and, to a limited extent, other information voluntarily provided. While State evaluation focuses on activity and capability assessments at each stage of the fuel cycle, some non-technical parameters, including

political, economic, security, legal, and administrative factors are taken into account as background context for the evaluation process.

3. Uses of open source information

Well-researched and reliable open source information can provide a valuable basis for comparison with other sources of information in determining the nuclear-industrial capabilities of a State, evaluating the internal consistency of declared nuclear fuel cycle programmes, identifying possible undeclared activities, and helping understanding historical issues, especially activities conducted prior to the implementation of safeguards. Closely related are the reviews to evaluate the consistency and completeness of additional protocol declarations, some elements of which largely depend on the availability and relevance of open source information: holdings and locations of source material; fuel cycle-related research not involving nuclear material; the manufacture of specified equipment; imports and exports of specified equipment and materials; and future plans for nuclear infrastructure development.

Focused open source collection and information-analytic reports examine specific issues in a State or on a safeguards topic. Transnational procurement networks and illicit trafficking of nuclear material may involve activities crossing the borders of several States. For other safeguards evaluation questions, in-depth research in the scientific and technical literature, often in non-English language sources, and close collaboration with technical experts are needed. The extent and complexity of such research and analysis are related to the history and sophistication of the nuclear infrastructure of a State, current and past fuel cycle activities, as well as to safeguards compliance and State transparency. Other focused collections and information analyses are used to prepare for complementary access and inspection activities and in some cases to evaluate findings from in-field verification activities.

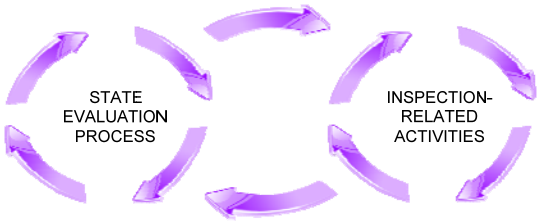

Figure 1. Relationship of the State evaluation process to inspection-related activities to be further integrated under the new conceptual framework (safeguards system that is fully information-driven).

4. Challenges and Responses

With the development of the State evaluation process and the implementation of additional protocols, the extent of information relevant to safeguards and the importance of its role have grown dramatically. The Agency has evolved from its traditional role as an auditor of declared nuclear material to an active evaluator of all information available to it. Since the 1990s, revelations of undeclared nuclear activities in several States under Agency safeguards, as well as advances in safeguards information management technologies, are supporting the development of safeguards that are fully information-driven as the future of modern nuclear verification.

The safeguards system will be 'fully information-driven' when all knowledge on a State is used to focus the appropriate objectives-based verification effort (rather than an effort based on prescriptive criteria), to achieve safeguards goals and to draw safeguards conclusions in the most effective and efficient way.

In this sense, knowledge is the cornerstone of the safeguards system; it constitutes the basis for planning safeguards activities, assessing results, and identifying follow-up actions required for drawing soundly based safeguards conclusions. This knowledge is acquired through the receipt of and/or collection and integrated analysis of all information available to the Agency such as State reports and declarations; information from verification activities; open sources; etc.

This holistic approach to safeguards implementation is applicable to all States and

is based on a comprehensive State evaluation and a State-level approach specifically tailored for each State. In the State level approach, verification activities are no longer conducted in a mechanistic, criteria-driven manner but rather are objectives-based and information driven, resulting in greater effectiveness and efficiency. Making the Agency's safeguards system fully information driven will require further evolution of processes already in place. [6]

After 15 years of continuous development, the Agency now possesses robust open source capabilities, both in acquisition and analysis. However, as outlined above, several challenges still remain. [7] Given that nuclear non-proliferation issues have risen to the top of the security agendas of many countries and, thus, are subject to wide debate, there has been a significant expansion in the volume of open source information, the number of sources and used the languages and formats. However, the presence of biased information and misinformation must also be taken into account in processing and analysing information for safeguards purposes. The problems are both exacerbated and ameliorated by developments in the information technology domain: web-based searching and extraction, and processing and archiving of growing amounts of information. In parallel, internal Agency requirements for open source research have been steadily increasing in order to support some 100 State evaluations conducted every year, and for other safeguards requirements.

Broader and deeper information retrieval relies not only on the identification of new sources, for example by developing access to information in non-official Agency languages, but also on the recognition of current limitations in available tools. For instance, only a relatively small fraction of the information is actually available through common search engines. Consequently, the Agency has acquired access to a number of subscription databases, mainly for scientific and technical information. Specialized software has also been acquired to locate and collect information that has not been indexed.

SGIM has pursued diversification of the information sources by accessing multilingual open source collections and databases, employing the linguistic skills of highly trained collectors and analysts, and utilizing a network of regional open source collection centres (set up with the help of Member State Support Programmes), among other measures taken to address information biases. In addition, while human resources constrain the volume of original-language articles that can currently be translated in-house, advanced machine translation tools are under evaluation.

The establishment of an integrated, service-oriented, analysis-friendly information architecture, as an effective base to collect, evaluate, analyse, structure, secure and disseminate safeguards information is an important element of enhancing information management capabilities. The increasing volume of information available makes it essential to continue increasing the efficiency of the collection and distribution process. Therefore, the use of SharePoint and Livelink technologies enables streamlined access to information collections, including open source "State files" supporting the State evaluation process as well as interdivisional communication and collaborative analysis. Furthermore, the Department of Safeguards development and implementation of a Geospatial Exploitation System (GES) in 2011 will further enhance the capability to undertake location-specific integrated analysis.

Finally, knowledge management has become the overarching objective for information management, if not for the overall management of verification resources. Key questions include: How do analysts handle the information they collect? How could retrieved information remain easily accessible? And how is continuity of knowledge propagated? Time is a factor that adds a critical dimension to the management of information. Not only do analysts need to know what they should know, but there will normally be no credible analysis of proliferation issues without securing the evolution of the "status" of a State over time. An issue that is considered resolved today, on the basis of information available, may become a major oversight in the future. Maintaining a historical "memory" of such evaluations in a structured way, adds further complexities to the management of information. In the long term, innovative solutions that provide a reliable basis for safeguards knowledge management will be necessary to guarantee that safeguards conclusions remain credible.

5. References

[1] Open source information is defined by the IAEA as information generally available to the public from various sources, such as government agencies, private entities, academic institutions and think tanks, the media, commercial subscription databases, scientific associations and commercially available satellite imagery. This information may be available in printed form (e.g. newspaper and journal articles, technical and scientific papers, reports and brochures), as well as electronic database entries and as Internet web pages.

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Note: A version of this paper was presented at “Global 2009: The Nuclear Fuel Cycle: Sustainable Options and Industrial Perspectives,” Paris, France, 6-11 September 2009, and published in Proceedings of Global 2009 as paper 9358.

High Resolution Radar Satellite Imagery Analysis for Safeguards Applications

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Abstract:

For monitoring nuclear sites, the use of Synthetic Aperture Radar (SAR) imagery shows essential promises. Unlike optical remote sensing instruments, radar sensors operate under almost all weather conditions and independently of the sunlight, i.e. time of the day. Such technical specifications are required both for continuous and for ad-hoc, timed surveillance tasks. With Cosmo-Skymed, TerraSAR-X and Radarsat-2, high-resolution SAR imagery with a spatial resolution up to 1m has recently become available.

Our work therefore aims to investigate the potential of high-resolution TerraSAR data for nuclear monitoring. This paper focuses on exploiting amplitude of a single acquisition, assessing amplitude changes and phase differences between two acquisitions, and PS-InSAR processing of an image stack.

Keywords: Synthetic Aperture Radar (SAR), TerraSAR-X, SAR Interferometry, in-coherent and coherent change detection, NFC signatures

1. Introduction

Space-based Synthetic Aperture Radar (SAR) is a technique for all-weather day and night observation. Compared to the European SAR-Sensors ERS and ENVISAT, the German SAR-Satellite TerraSAR-X has improved the available spatial resolution from 20m to 1m, which allows the identification and change monitoring of smaller buildings and even to identify structural features on them. However, as SAR images are subject to microwave scattering phenomena and have a different imaging geometry than optical imagery, they are challenging to interpret and analyse.

In this study two series of radar scenes (stacks) covering the Forschungszentrum Jülich and adjacent areas are collected, coregistered and analysed in order to investigate the suitability of radar signals for nuclear monitoring tasks. Additionally with these 2 image stacks, several interferometric pairs of the same area are acquired and processed, fully exploiting the wide variety of TerraSAR-X acquisition parameters. The results will show the effects of the acquisition parameters for the recorded image and serve as a decision support for future acquisitions.

Moreover, the study includes the application of SAR interferometry (InSAR) for the determination of building geometries. Multi-temporal in-coherent techniques are applied to detect possible building-shape, building-deformation and (de-)construction activities. Digital Elevation Models derived from TanDEM-X and Worlview-2 optical data are available for validation purposes.

2. TerraSAR-X Mission and Data

This document will concentrate on data acquired by the German SAR-Satellite TerraSAR-X, but data recorded by other SAR-Satellites like the Italian COSMO-Skymed, the Canadian RADARSAT 2 (both X-Band), the ERS-1/2 & ENVISAT satellites (C-Band) operated by ESA, and the Japanese ALOS-PALSAR (L-Band) can be exploited in similar ways. Each wavelength shows distinctive assets and drawbacks, however, short wavelengths like X-Band (3.1 cm) generally offer the best resolution.

The TerraSAR-X satellite, Germany's first national remote sensing satellite, was implemented in a public-private partnership between the German Aerospace Centre (DLR) and EADS Astrium GmbH. It was launched in June 2007 and carries an advanced high-resolution X-Band Synthetic Aperture Radar using the active phased array technology to acquire images in various modes.

TerraSAR-X Overview	
Antenna length:	4.8 m
Weight:	1.230 kg (including payload mass 400 kg)
Orbit:	514 km
Inclination:	97.4°, sun-synchronous
Repeat cycle:	11 days
Launcher:	Dnepr 1 (former SS-18)
Launch:	15 June 2007, 4:14 h (CEST) from Baikonur, Kazakhstan
Life time:	5 years (minimum)
Radar Frequency:	9.65 GHz
Transmit Bandwidth	100 / 150 MHz nominal 300 MHz experimental
Polarization:	HH / VV / HV / VH
StripMap Mode: [Range × Azimuth]	Resolution: 3 m × 3 m Scene Size: 30 km × 50 km
SpotLight Mode: [Range × Azimuth]	Resolution: 1 m × 1.5 m...3.5 m Scene Size: 10 km × 5 km...10 km
ScanSAR Mode: [Range × Azimuth]	Resolution: 16 m × 16 m Scene Size: 100 km × 150 km

Table 1: TerraSAR-X Mission parameters, modified after Buckreuss [1].

To cover wide areas, the satellite can be operated in the ScanSAR Mode, recording a swath width (image width on the ground) of 100 km and a length of up to 150 km. The higher the resolution of the selected mode, the smaller the footprint (imaged area on the ground), leading in the end to the High-Resolution Spotlight Mode width a scene extent of 5 x 10 km and a resolution of up to 1 meter. Offering the best resolution, solely data recorded using the High Resolution Spotlight Mode is used for the presented work. Figure 1 shows the amplitude information of a single High-Resolution Spotlight TerraSAR-X scene acquired over Juelich Research Centre, parts of Juelich City and Hambach and Inden opencast pits and surrounding agricultural and forest areas. The amplitude information is presented in range (horizontal) and azimuth (vertical) coordinates.

All SAR acquisitions have in common that the image coordinate-system is a cylindrical one, as the SAR-sensor is measuring the distance to the backscattering feature on the surface. The system immanent properties are causing geometric "distortions", which can already be exploited as shown in chapter 3.1.The different acquisition modes and their parameters in detail are publicly available on the TerraSAR-X Science Service (http://sss.terrasar-x.dlr.de) [2].

Figure 1: Example of the amplitude information of a single High-Resolution Spotlight TerraSAR-X scene, presented in range (horizontal) and azimuth (vertical) coordinates.

3. Processing Methods and Examples

Each radar pixel consists of two "layers" of information:

- Amplitude:
  The energy backscattered by one resolution cell.
- Phase information:
  A measure for the distance between antenna and scatterer or changes of the scatterer's structure (measured modulo 2π).

Every layer contains different information and different methods have to be applied. The following sub-chapter will briefly present these methods and application examples and how they can help to identify visible indicators related to nuclear fuel cycle facilities and processes. In detail, the possibilities offered by a single image, two images and an image stack will be exploited.

3.1. Exploiting amplitude of a single acquisition

A single radar acquisition offers, besides the "imaging" of the area or geodetic localisation of certain features as shown in Eineder [3], the possibility to retrieve certain information of buildings, exploiting the SAR-specific geometric imaging properties like shadow, layover and foreshortening, methods and examples are demonstrated in Wegner [4].

Depending on the chosen incident angle for the radar acquisition and the ratio between the height and width of the monitored building, different formulas have to be applied. Both information can be retrieved by measuring the length *S* (in range) of the shadow generated by the building and the length *R* (in range) of the roof in image coordinates and transforming them to metric values using the image resolution (Figure 2).

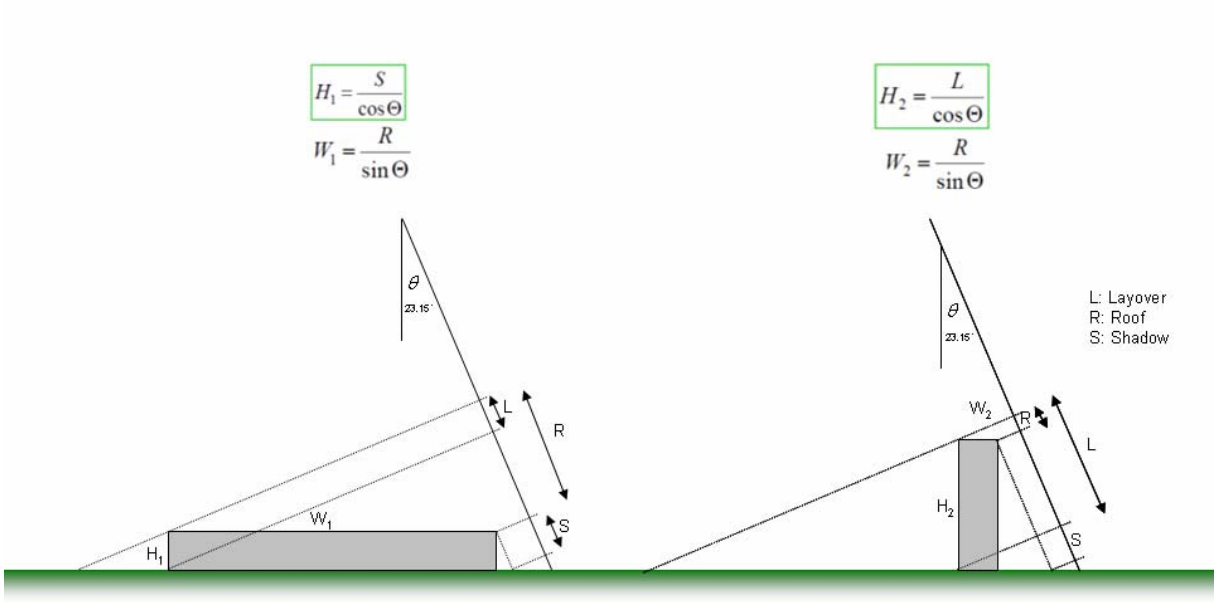

Figure 2: Geometric relation in SAR-imaging of buildings, showing the trigonometric relations of real height and width to SAR-image layover and shadow.

3.2. Amplitude changes between two acquisitions

Adding a second SAR acquisition, acquired with the same set of parameters but at another time, greatly enhances the possibilities of information extraction, including all sorts of temporal changes. Just by comparing the amplitude of the two images, changes of buildings, infrastructure, mining activities and certain reflecting objects like cars, trains or containers can be identified (Figure 3).

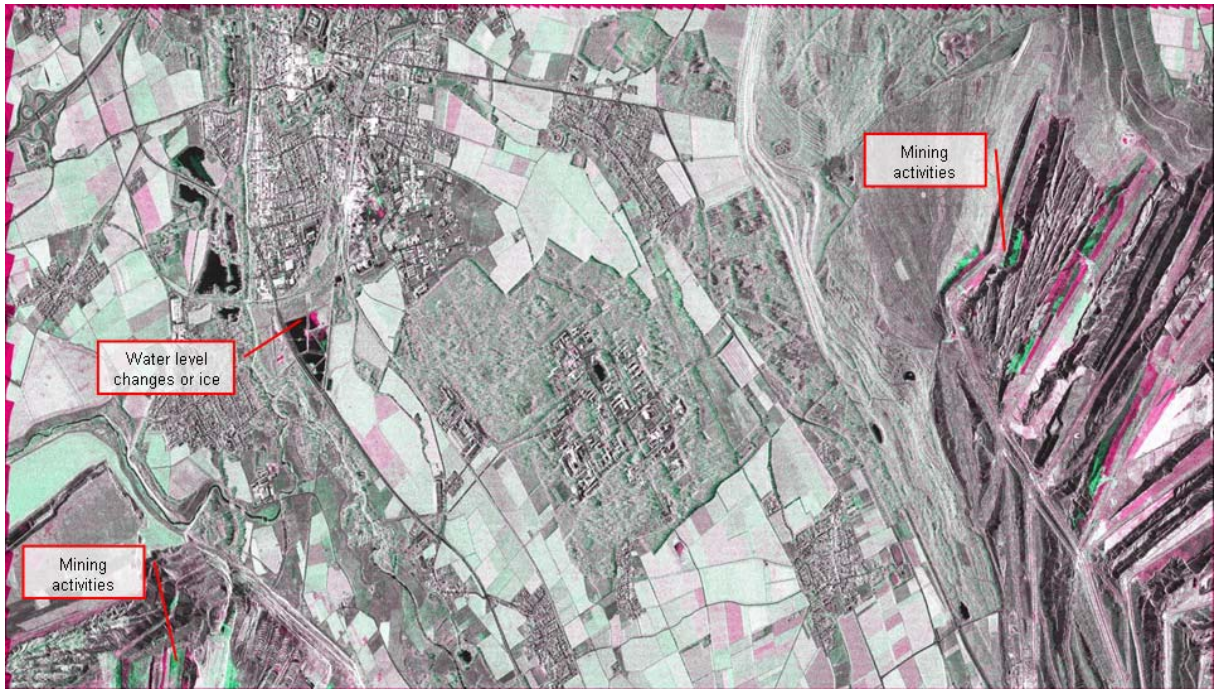

Figure 3: Amplitude changes between two SAR-Acquisitions (temporal distance 22 days), showing mining activities and water level changes, (red: brighter in 1st image, green: brighter in 2nd image).

Metallic objects, especially like containers, ships, cars, wagons, etc. show strong backscatter and are therefore easily identifiable in radar acquisitions. Figure 4 shows the relocation of containers, between the two acquisitions with a temporal separation of 33 days and mining activities at an opencast pit.

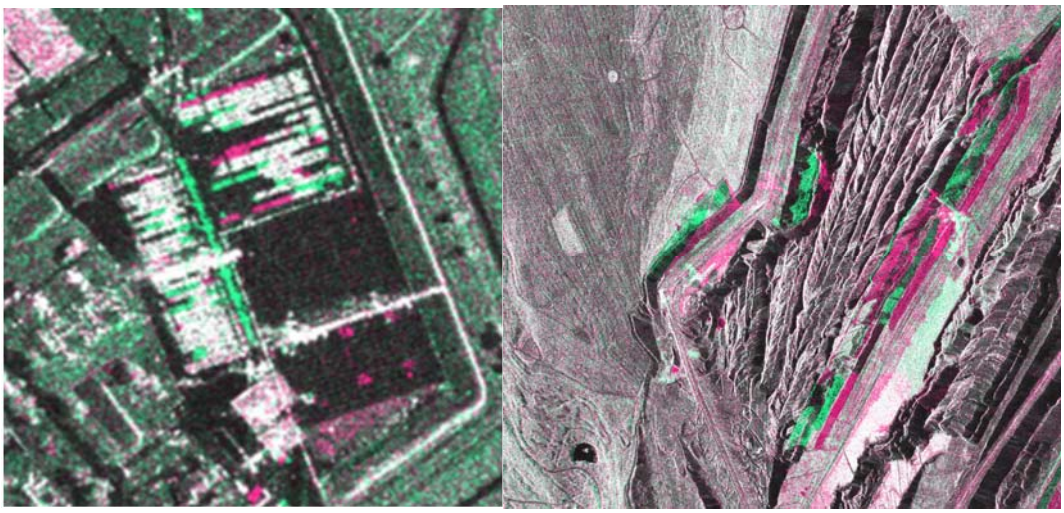

Figure 4: Amplitude changes showing relocation of containers (left) and mining activities (right), (colours like in Fig 3, temporal distance 33 days, respectively 22 days).

3.3. Phase differences between two acquisitions

If two radar images, acquired using the same set of parameters, are available, analysis is not restricted to comparing the amplitude, but the phase information recorded for each pixel can be exploited as well. After correcting for topographic effects using digital elevation models (DEM's), each pixel contains mainly information about deformation, deviations from the used DEM and atmospheric path delays due to propagation effects of the radar signal caused by changes in water vapour and TEC content of the ionosphere. The path delays have to be treated as errors, but both deformation and DEM-deviations can be used to detect and measure buildings and their changes and even shallow underground activities by detecting subsidence caused by them (Figure 5).

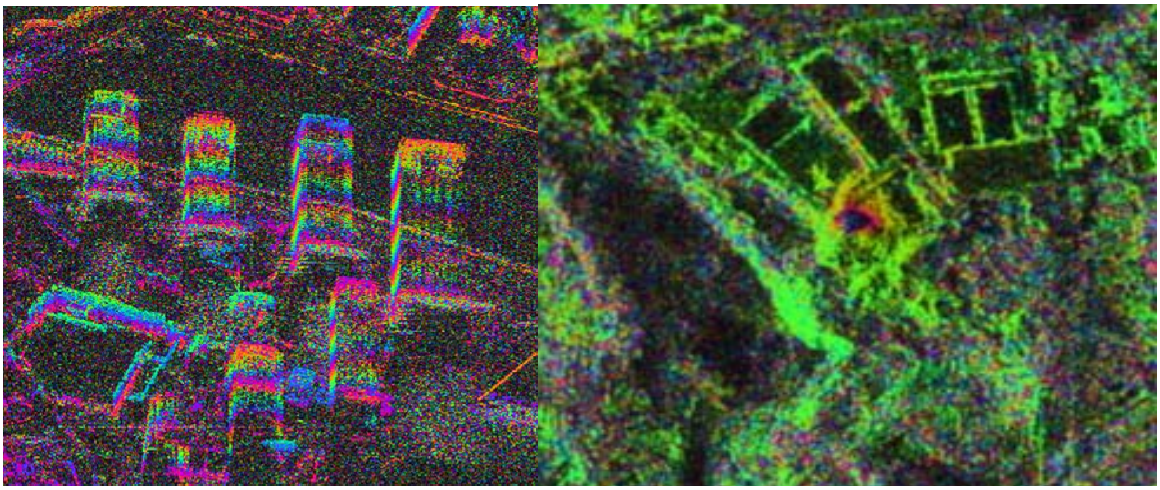

Figure 5: Left: Interferometric height measuring shown at skyscrapers in Tokyo, one colour-cycle corresponds to 43.8 m height, Right: Interferometric deformation measurement at Campi Flegrei, showing ~1 cm uplift over 11 days due to fumarolic activities, presented in Minet [5].

3.4. PS-InSAR processing of an image stack

Persistent Scatterer Interferometry (PS-InSAR) offers a very accurate measurement for detecting deformations in the scale of millimetres per year. However, for a reasonable application, an image stack of at least 10, or better more images are required. The method is based on restricting the processing to a set of targets (up to several millions in TSX processing) with a strong and stable signal. By creating a network between these "persistent scatterers", measuring of relative displacements and height differences between each pair of points and finally integrating the whole network with respect to a selected, stable reference point, propagation effects of the SAR signal can

be removed and small deformations can be detected and monitored. Another result of PS processing is the difference of each scatterer from the used DEM. Since DEMs rarely include buildings, the retrieved values allow the reconstruction of building, as shown in Bamler [6].

Monitoring the same area from different aspect angles may also solve the problem of missing walls of buildings due to the already mentioned shadow effect. By combining these results, full 3D reconstructions of buildings and building features [7] can be performed (Figure 6).

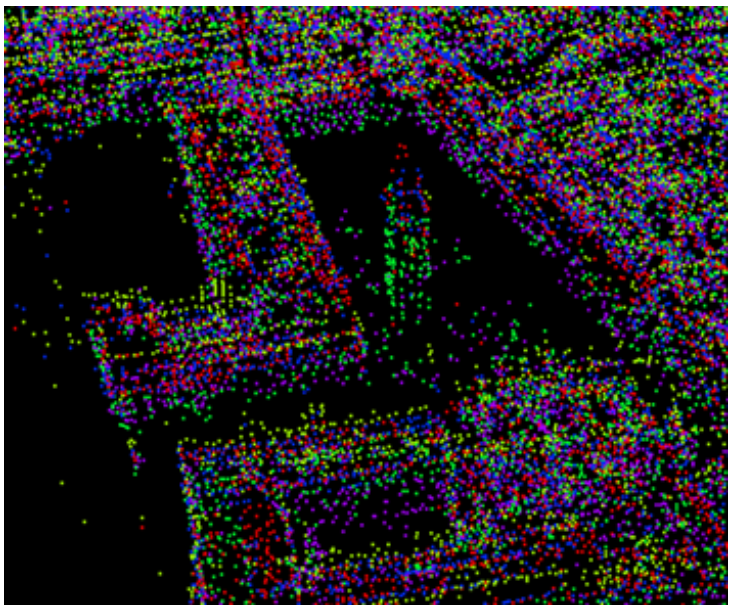

Figure 6: Combination of 5 PS-InSAR stack reconstructing the Campanile and Piazza San Marco in Venice, each colour representing one observation direction, processed by Hanisch [8].

By now, the image stacks acquired for the presented project are not large enough to be processed, but data acquisition and collection is ongoing to prepare two full image sets.

4. Digital Elevation Models

DEMs, generated from miscellaneous data (SRTM, TanDEM-X, Worldview-2) that were acquired at different dates, allow a comparison of larger changes of the earth's topography. Especially the mining progress in opencast pit and the volume of stockpiles waste rock can be estimated by comparing different DEMs. The swell factor of the dumped waste rock and even the amount of the produced raw material can be estimated (Figure 7).

5. Conclusions and Next Steps

Data acquired by SAR-Sensors, especially space-borne high-resolution sensors like TerraSAR-X, poses several possibilities to detect and monitor changes of the Earth's surface and man-made infrastructure. The possible results of the different methods, starting with a single acquisition up to a complete image stack were shown using High-Resolution X-Band SAR-Data acquired by the German SAR-Satellite TerraSAR-X. The independency of lighting and atmospheric conditions recommends this sensor for surveillance applications. However, the 11-day repeat-cycle has to be kept in mind, if an interferometric processing should be applied.

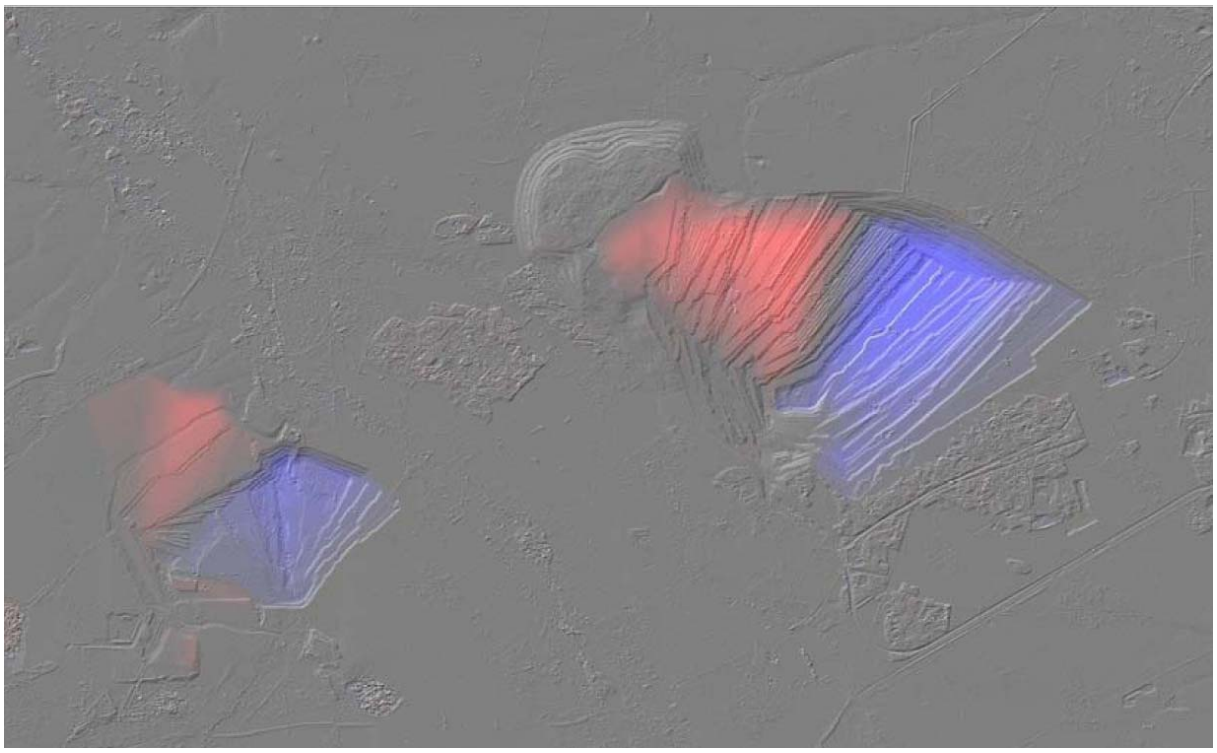

Figure 7: Mining Progress at Inden and Hambach opencast pits between 2000 (SRTM-Data) and 2010 (TanDEM-X data), red indicating waste rock deposition and blue indicating material extraction

It was demonstrated, that a general impression of the covered area, highly accurate localisation of special features and information of building heights can already be retrieved from a single acquisition. Two images permit the monitoring of a wide variety of temporal changes, such as changes of buildings, infrastructure or deformation of the Earth's surface, and also enhance the height estimation of buildings. Using a stack of ten or more acquisitions of the same area with identical acquisition parameters, the PS-InSAR method allows a complete reconstruction of the 3-dimensional structure of buildings, including large scale features and also the measurement of slow deformations. Additional to the abovementioned method, a comparison of high-resolution DEMs, acquired at different times proved to provide valuable information.

The next steps will focus on analysing and interpreting radar signatures from the image stack acquired over Juelich Research Centre, by also considering information collected at the ground.

6. Acknowledgements

The work is funded under task D1657 of the German Support Programme to the IAEA. The authors want to thank Dr. Pablo d'Angelo of the Department for Photogrammetry and Image Analysis (Remote Sensing Technology Institute, DLR) for organizing the Worldview-2 stereo data and calculation the DEM.

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Improvement of Design Information Verification capacities by specific implementation of available geophysics

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Abstract

Efficient safeguards (SG) implementation at complex installations is critical for the Agency to better fulfil its mandate in providing assurances of the absence of diversion of nuclear material at declared facilities. Structures might be difficult to access, making the Verification of Design Information (DIV) or the Complementary Access (CA) a real challenge to inspectors. Geophysical techniques such as ground penetrating radar (GPR), authorized by IAEA for inspection in 2006, could help inspectors to get a larger and better comprehensive view of an installation and its activities. They could provide information by mapping infrastructures and their content and any accessible geophysical anomalies.

After an analysis of SG Inspections targets and operational context, two families of alternatives and complementary appropriate techniques were identified. Inside a building, tools such as GPR (electromagnetism properties), Thermography (thermal anomalies), Impact Echo or Ultra Pulse Echo (sonic parameters) will help verifying the structure of a wall, its contents such as tubes, as well as the structure of a slab. It will also enable access to the information about the space beyond wall or slab and its content such as void and metallic or plastic features. In addition, activity of features such as pipe beyond wall will be addressed by their temperature. Outside a building, GPR, Magnetism (magnetic parameters) and Active Seismic (velocity of elastic waves), will provide information on the existence or absence of underground structures or features such as tanks, pipes, sewers, … The combination of these complementary and alternative techniques will ensure more reliable results.

The selection of such geophysical techniques, the analysis of their capacities and limits regardless to IAEA objectives, and their implementation mode were analysed by CEA as part of the French Support Programme, with the support of IFSTTAR, former Laboratoire des Ponts et Chaussées (LCPC) bringing its expertise in NDT and subsurface survey, and its geophysical test site, with recent involvement of EOST (Ecole et Observatoire des Sciences de la Terre) for magnetism.

The geophysical signatures of anomalies for DIV inspection objectives were analysed for different inspection contexts. GPR was implemented in a first step. Other preselected techniques could be soon implemented.The programme produced specific basic and continuous training program, field examples catalogue to help inspectors, standard acquisition and processing configurations and possibilities of software simplification. The program mainly aims to get useful data from inspection. Recent deeper analysis of an appropriate field approach lead to usage of lighter techniques before the slower one.

Beyond technical approach, one remaining challenge is the full integration of such particular and new techniques requiring specific skills and usage in the IAEA, leading to all processes adaptation, from identification of appropriate usage identification during inspection preparation to specific training, maintenance and data handling as well as processing and interpretation processes.

Despite the issue to manage low frequency usage, some limitations and the requirement for a specific training for core of inspectors, these additional tools based on available equipment, gives to IAEA the capability to better address future challenges by getting information actually not accessible.

Keywords : DIV, complementary access, safeguard inspection, undeclared activities, geophysics

Some operational methods to analyse radar images

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Abstract:

The CEA developed an interferometric software package called CIAO (Chaîne Interférométrique rAdar Opérationnelle). The objective is to obtain an automatic tool for sites detection and monitoring. Coherence images and differential interferograms are computed using CIAO to highlight changes between at least two radar images acquired on a same site. Those results are georeferenced to make relevant analysis.

As CIAO is constantly improved, the four latest developments are described.

- *Due to the acquisition geometry, the perception of a SAR image is different compared to an optical system. Distortion effects like layover, foreshortening or stretching of the back slopes make the image difficult to interpret, especially for man-made structure. For example, buildings are often not rectangles but only one bright stripe or bright spot on the images. This renders visual interpretation of SAR imagery less intuitive than optical imagery and usually an experienced analyst is required for the task. To facilitate the interpretation of SAR images, a georeferencing capability was added to CIAO. If the analyst has an optical georeferenced image and a radar image acquired on the same area, he can associate visually a region of the radar image to the corresponding area in the optical image.*

- *It is possible to use as input of CIAO images acquired by the satellites ERS, ENVISAT, TerraSAR, Cosmo-SkyMed and ALOS. Therefore an analyst can take advantage of the different physical properties depending on the band used by the satellites.*

- *In some cases, atmospheric effects can decrease the measurement precision of small ground movements. If atmospheric data are available at the acquisition time, it is possible to compensate those effects with an atmospheric module which has been added to CIAO.*

Keywords: CIAO; CEA software tool, change detection; interferometry; atmospheric compensation; Permanent Scatterers

1. Introduction

This paper presents the latest improvements of the software package CIAO developed by the CEA for sites detection and monitoring using radar imagery that permits all weather, day and night collection capabilities.

Computing the data called the *amplitude image*, radar images can help the investigation of areas with frequent cloud coverage, night acquisition, snow covered areas where radar can penetrate the snow,… This data can be difficult to interpret because of distortions due to the acquisition geometry. So a georeferencing capability was added to CIAO. Another useful data to detect human activities is the *coherence image*. The analyst first collects the radar images for a given area (possibly from different satellites) and then uses the CIAO software to identify and compute possible coherence

images. Using high resolution radar images, the deformation of a particular area can be computed. This measurement can indicate underground buildings. If the atmospheric data are available at the acquisition time, it is possible to compensate those effects to improve the accuracy of the deformation mesurement. Considering the latest high resolution X-band satellites, the detection of Permanent Scatterers in a temporal series of radar images can indicate human activities.

2. Amplitude Image : need of a georeferencing tool

One of the interests of radar images is the all weather and time capability. However using radar images as stand alone data could be difficult because of the distortions due to the geometry acquisition. There are three kinds of distortions: layover, foreshortening and shadow [1]. Furthermore, when analysing the amplitude image, the position of North, South, West and East can change from an image to an other. So, change detection between two radar images (figure 1), is simplified if the data are georeferenced. CIAO takes into account the physical properties of ERS1-2, ENVISAT, TerraSAR and Cosmo-SkyMed. Figure 1 is an example of two Cosmo-SkyMed images acquired on the nuclear site of Olkiluoto (Finland), which hosts a nuclear facility under construction during the acquisition period. Human infrastructures can easily be detected but they are difficult to identify. There are a lot of bright stripes and bright spots corresponding to the interaction between the wave emitted by the satellite and the man-made structures.

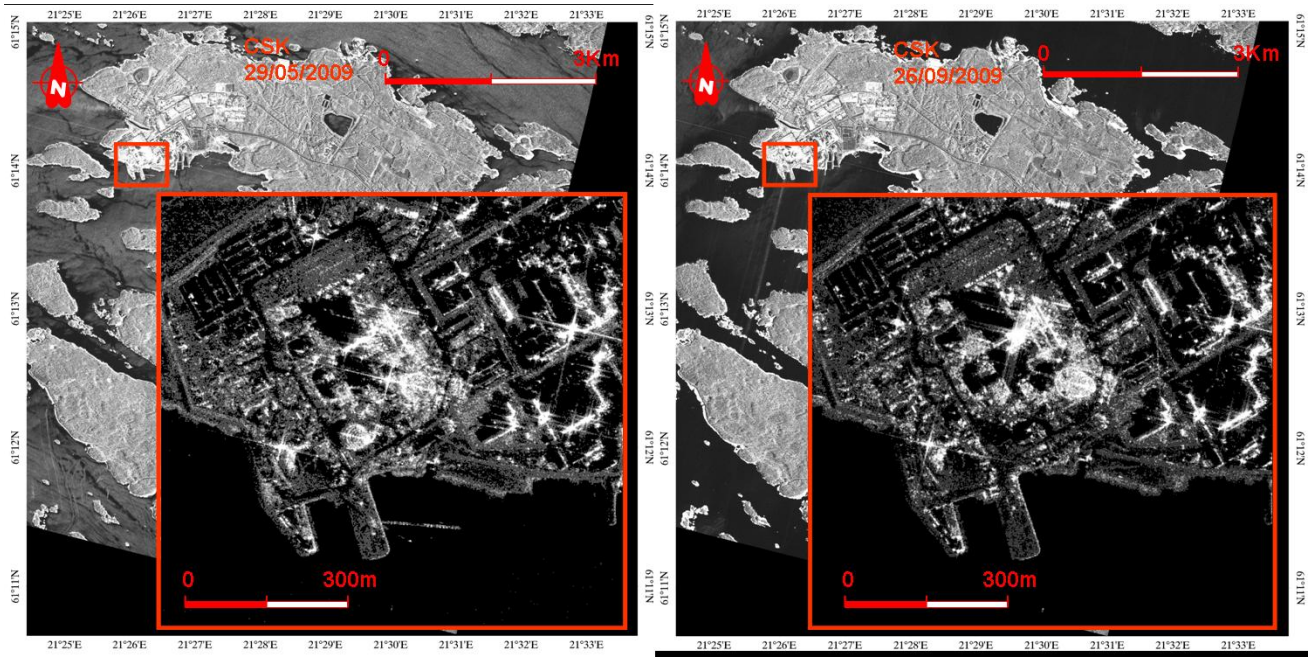

Fig. 1: Two Cosmo-SkyMed amplitude images after georeferencement. Both images are in the same geometry. The left one was acquired on May 29th 2009 and the right one on September 26th 2009. The red rectangle is a zoom on the nuclear site of Olkiluoto (Finland).

3. Coherence image

A coherence image is computed with two coregistered radar images. Dark areas in the coherence image indicate changes between two images and the bright areas indicate possible infrastructures which did not change between the two image acquisitions. The coherence image can also be used for site monitoring [2]. High resolution radar images improve significantly the number of areas detected as changes. The benefit of the image presented on figure 2 is the precise localization of changes. The site presented is the same as on figure 1 (the nuclear site of Olkiluoto). So the idea is to use the amplitude image of those two images and the coherence image. The oldest amplitude image is in the red canal, the latest amplitude image is in the green canal and the coherence image is in the blue canal. Thus in the area detected as changes in the coherence image, there is no blue. Thus, structures present only in the oldest image are red and structures present only in the latest image are

green. An example is presented figure 2. Red and green structures are clearly identified. The yellow area corresponds to stable structures.

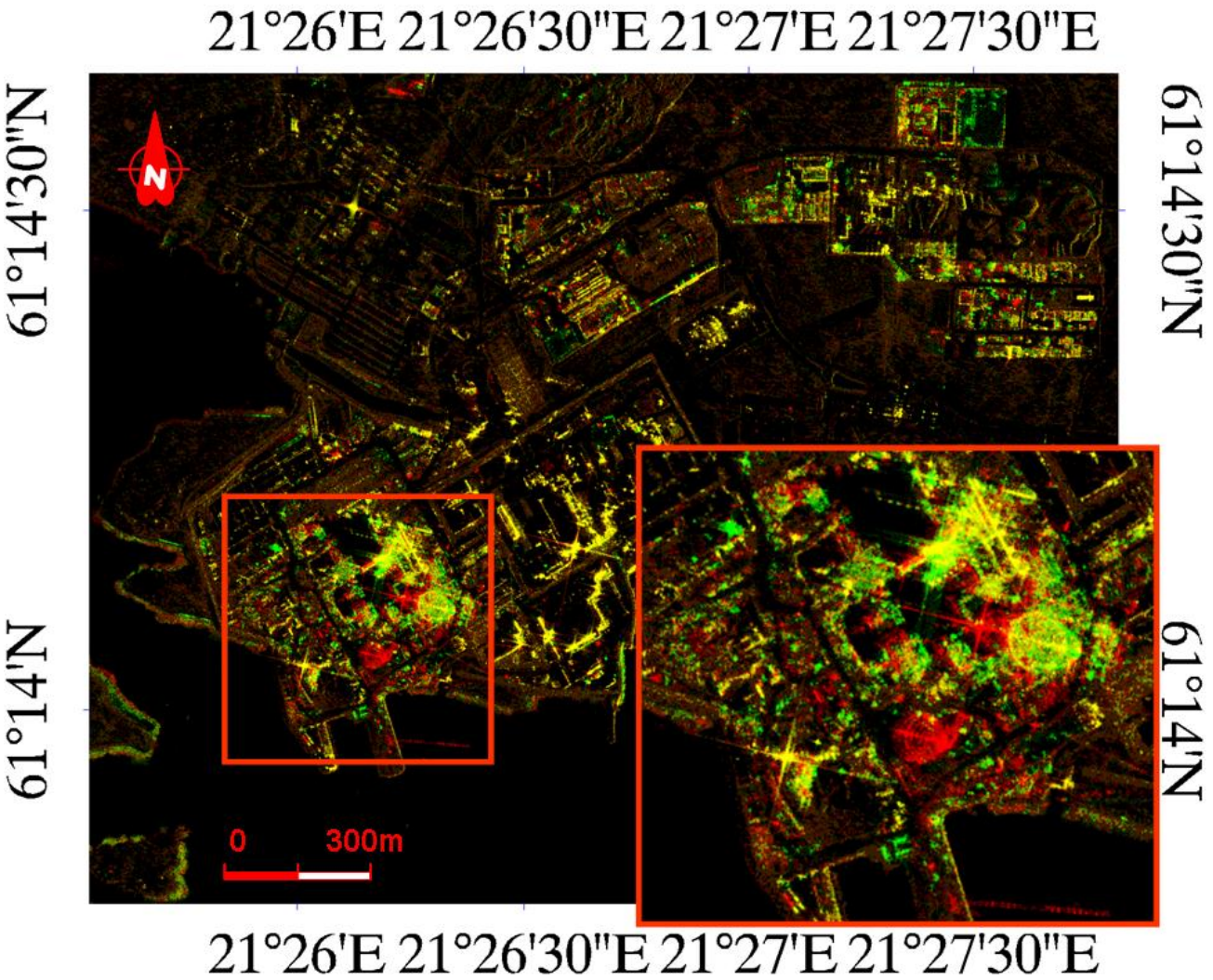

Fig. 2: Colour composition: the red canal corresponds to the amplitude of Cosmo-SkyMed image acquired on May 29th 2009, the green canal to the amplitude of Cosmo-SkyMed image acquired on September 26th 2009, the blue canal to the coherence image computed with both radar images. Red structures are only present on the first image and green ones on the second image. Yellow corresponds to stable area.

4. Deformation : atmospheric compensation

From the phase difference of radar images acquired at different dates, CIAO computes Differential Interferometric Synthetic Aperture Radar interferometry (DInSAR). It yields maps of ground displacement. Interferograms can be affected by coherent noise like atmospheric effects. To improve the measurement of ground deformation two algorithms were implemented in CIAO. Considering a DInSAR, the idea of those methods is to simulate an “atmospheric interferogram” using meteorological modelling to estimate the tropospheric radar delay. The first algorithm, dedicated to the ENVISAT images, uses the Mesoscale Meteorological Model, MM5, [3] and multispectral Medium Resolution Imaging Spectrometer Instrument (MERIS) images (the MERIS system is on board the ENVISAT platform [4]). The second one uses the Weather Research and Forecasting (WRF) model to compute the atmospheric delay. WRF is suitable for a broad spectrum of applications across scales ranging from meters to thousands of kilometers. In this paper we present a result obtained on Tiberias Lake region [5]. Two ENVISAT images were acquired: on January 4th 2004 and the other on March 14th 2004. Assuming that no seismic signal occurred between the two images, interferogram computed

with CIAO is compared to the atmospheric one. The figure 3 shows profiles of the CIAO interferogram (thin line) and of the interferogram simulated using the first algorithm. Those two profiles superimpose well and have approximately the same amplitude (about 1 rad).

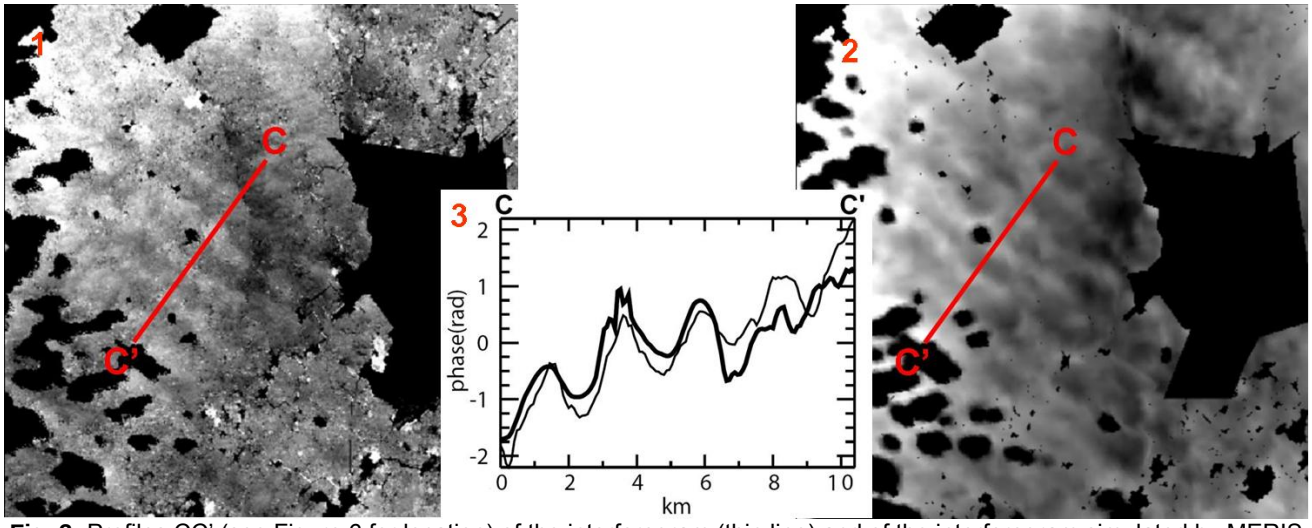

Fig. 3: Profiles CC' (see Figure 6 for location) of the interferogram (thin line) and of the interferogram simulated by MERIS and MM5 (thick line). The profiles have been averaged on a 10 km long segment perpendicular to CC'. Oscillations, 2.25 km in wavelength, superimpose well and have approximately the same amplitude (about 1 rad).

This algorithm that estimates the atmospheric phase delay should significantly improve the potential of SAR interferometry for the measurement of tectonic deformation, in particular interseismic deformation, where strong atmospheric effects are currently a major source of limitation. It removes about 43% of the atmospheric signal

4. Permanent Scatterers technique

Permanent Scatterers, or Persistent Scatterers (PS), are stable ground reflectors. Their reflection properties remain stable during long time periods, so their movements can be monitored with a better accuracy than other points, which would be influenced by decorrelation noise. Indeed, the phase of a pixel containing a PS is determined mainly by the own phase of the PS, rather than by the phase components of other non-PS reflectors contained in the pixel. In other words, a PS has a dominant behaviour in a resolution cell.
In order to detect PS, we need a time series of stackable radar images. Thus, all images have the same range and azimuth resolution and every single pixel represents the same ground area in all the images. PS detection is then performed through a pixel by pixel analysis, studying amplitude variations of pixels along the time series. Amplitude stability is linked to phase stability when Signal to Noise Ratio (SNR) is high [6]. This hypothesis is validated because PS have very high amplitudes values. The amplitude dispersion index is defined as follows [6]:

$$D_p = \sigma_p / \tilde{A}_p$$

where

- σ_p is the standard deviation of the pixel amplitude along the time series,
- $\tilde{A}_p$ is the average value of the pixel amplitude along the time series.

The more the amplitude is stable, the more the dispersion index is small. Thus, to determine if the pixel is a PS candidate or not, the amplitude dispersion index is compared to a threshold, typically 0.25. If the dispersion index of the pixel is lower than the dispersion threshold, the pixel is detected as PS candidate.
The idea is to use the map of dispersion to detect human activities. Indeed, PS (in red on figure 4) can indicate typical human structures. The dispersion data presented figure 4 was computed with a time series of twelve coregistered TerraSAR images. Those images were acquired on the Serre Ponçon dam in the Southern French Alps on the verge of the two departments Hautes-Alpes and Alpes de Haute Provence. The dam is well identified and appears in red. East of the dam height variation of water is visible on the banks of the lake. West of the dam, high tension power lines appear as lines on

the lake: it is due to the high dispersion. This example shows the potential of the dispersion image to detect human activities.

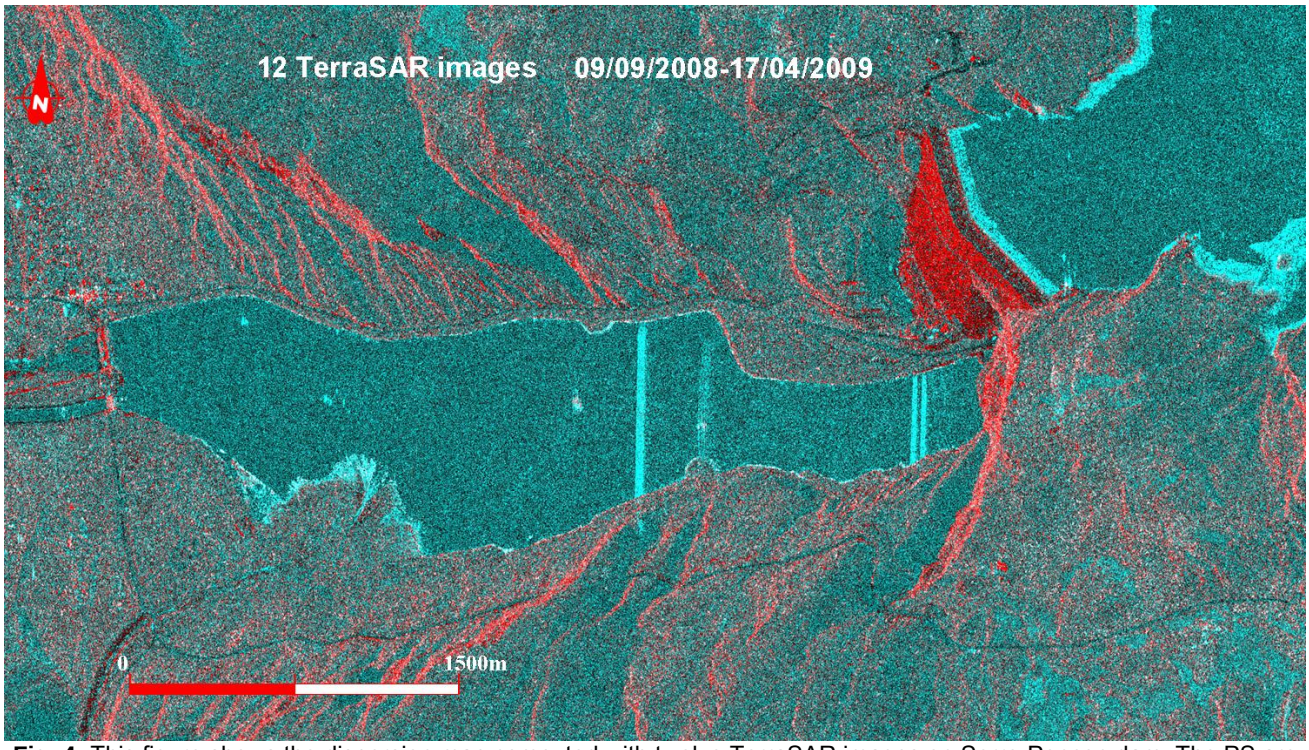

Fig. 4: This figure shows the dispersion map computed with twelve TerraSAR images on Serre-Ponçon dam. The PS are red. The first image was acquired on September 5th 2008 and the latest on April 17th 2009. The dam clearly appears in red.

After this PS detection step, a displacement parameter estimation can be performed. Such methods are described by Ferretti & al.[6] or Bert M.Kampes & al.[7]. It is possible to evaluate linear and non-linear displacements rates along the Line Of Sight (LOS) of the satellite. Digital Elevation Model (DEM) errors can also be estimated.

4. Conclusions

The techniques presented in this paper show that radar data can be useful for change detection. CEA develops a software package called CIAO whose goal is to obtain an automatic change detection tool using radar data. Some work remains to achieve this objective. One of the challenges is to obtain an automatic module for registering radar images acquired by different satellites with optical images.

5. Acknowledgements

The work presented is the result of the radar team of the CEA, Béatrice Pinel-Puysségur, Guillaume Quin and Pierre Duperray.

6 Legal matters

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Open Source Geographic Information for Safeguards Analysis

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Abstract:

In this era of user-generated Web content, geographically referenced information is being published to open sources at an astounding rate. One might conceptually understand these data as the product of a distributed, decentralized sensor network capable of detecting the geographic signals of nuclear proliferation. Within an information-driven safeguards regime, these data, often created and shared by common citizens, can be invaluable to the detection of undeclared nuclear activity. Such information, however, is often overlooked and underutilized because, at present, no tools exist to systematically and efficiently extract and utilize these data. This paper describes an ongoing project that seeks to enable safeguards analysts to efficiently and effectively use open source geospatial information by leveraging web-based information technologies in novel ways.

While a great deal of geospatial data are published in well defined, easily detectable formats, most data are unstructured, heterogeneous and complex. Geospatial and domain-specific ontologies can be used to detect and convert these data into usable and semantically interoperable formats that can be effectively incorporated into an analyst's work. Working closely with safeguards analysts and other stakeholders to establish high-level requirements and derive use cases ensures that these tools are integrated into analysts' existing workflow for efficient use and high adoption.

Keywords: Open source; safeguards; geospatial; GIS; analysis

1 Introduction

Today's Internet is characterized by numerous interactive features that provide a plethora of avenues for user-contributed content. Ubiquitous cell and smart phones usually combine camera, web browser, global positioning system (GPS), and other tools that permit the person on the street to upload text, photographs, and video to any number of blogs, social media outlets, news networks, and other online repositories in near-real time. Growing numbers of tablet computers further swell the ranks of highly mobile, amateur and professional web authors.

Much of this user-generated content is geospatially referenced. For example, modern digital cameras embed spatial coordinates by default, tweets (140-character Twitter messages) and Facebook updates allow users to "geotag" their location. Geospatial information can simply be entered manually, obtained from cell tower triangulation, or precisely derived from GPS.

And while data in social media streams are unstructured in the extreme, geospatial information can very often be systematically extracted. By way of an example, one of us gives his Twitter location as "Usually ABQ," reflecting an approximate location due to frequent travel for work. Twisst, a Twitter-based service that notifies users of daily flyover times of the International Space Station [1], manages to recognize "ABQ" as the airport code for the Albuquerque International Airport and derived the correct time zone and correct location to within 8 miles. Twisst then sends out a personalized tweet

[1] Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000. SAND # 2011-3018C.

when an ISS flyover is imminent. Still others have demonstrated that an individual's location can be derived from implicit geographic information by analyzing regional colloquialisms and high-level topics within the text of Twitter posts [2].

Importantly for the purpose of this study, some of this web content may accidentally, incidentally, or purposefully include information on nuclear facilities, materials, and perhaps even proliferation activities. However, such information is often overlooked or not used because the scripts to extract these data must be hand-crafted. Even when a well-defined Application Programming Interface (API) is available, a solution must be constructed for each data source. In order to make this information available to safeguards analysts, web-based technologies need to be leveraged in novel ways so that the end user need not rely on web programming expertise.

All of this points to a large and growing body of data that has the potential to contain geospatially referenced, safeguards-relevant information. Within an information-driven safeguards regime, these data, often created and shared by common citizens [3] can be invaluable to the detection of undeclared nuclear activity. One might conceptually understand social networks as a distributed, decentralized sensor network capable of detecting the signals of nuclear proliferation, often with geospatial metadata. Unfortunately, these data are hidden in a forest of innocuous information and as such, are often overlooked and underutilized because no tools currently exist to systematically and efficiently extract and make use of these data.

Recently, articles discussing nuclear proliferation detection using geospatial data have focused on the use of aerial and satellite imagery for change detection analysis [4], [5]. This has led the way to the systematic use of satellite and aerial imagery within the safeguards community as one way of identifying undeclared nuclear activity [6]. Other geospatially referenced open source information, such as ground level images from tourists and visitors, "crowdsourced" map data, and geospatial references in blogs or discussion wikis are a resource that have not been systematically analyzed to determine their usefulness in safeguards analysis.

This research was designed to survey current geospatial resources on the open Internet and examines the feasibility of providing geospatial tools to analysts who do not have a high level of GIS or web programming fluency. Therefore, the hypothesis of this work is that, by enabling safeguards analysts to efficiently and effectively extract and utilize geospatially referenced information from the Internet, these analysts will more often use these data to produce more complete and context rich analyses.

2 Methods

Several facets to the research have been identified for this project: (1) The identification of current open source tools with the potential to assist analysts in extracting and managing geospatial data; (2) an examination of the growing number of geospatial data types and the spatialization of typically non-geographic data like photographs; and (3) a test case to demonstrate of the usefulness of these types of open source geospatial information.

2.1 Tools assessment

To guide the assessment of the tools available to achieve these goals, several requirements were established for the development of a safeguards toolset. First, the tool should be available at low or no cost to the end user. This requirement led to the examination of existing open source software that could be used, modified, and re-distributed free of charge and copyright restriction. Moreover, by leveraging open source software, existing capabilities need not be reinvented, thus saving significant development time.

Second, the developed tools should integrate easily into analysts' existing workflows. This will help to ensure that the tools can be used widely among analysts by lowering the threshold of adoption. This also requires that tools have an intuitive design and functionality, refined user interface, and that users are closely involved in the development. This further requires that these tools be compatible with existing software and data.

Third, these tools must be extensible and thus adaptable to different analysis requirements, individual preferences, and future changes in workflow.

One tool that has been identified as a candidate for further development as a safeguards-specific tool is Zotero, a reference management software used to collect, organize, cite and share electronic research sources [7]. Because the workflow of safeguards analysis has a clear correspondence to the research process in which references are collected, tracked, and organized, and reports are generated, Zotero appears to be a valuable starting point for development. Also, because Zotero is free, open source, and extensible, it meets all of the established requirements for this project. Currently, Zotero is deployed as an add-on for the Firefox web browser and therefore integrates into one of the primary tools of safeguards analysts—the web browser (Zotero is also being developed for use with other browsers and as a standalone tool.) Additionally, several modules are available that extend the core functionality of Zotero including word processor integrators and, of particular interest to this work, mapping capabilities.

Zotero Maps, an extension to the core Zotero program, identifies and geocodes place names within documents managed by Zotero. Figure 1 was created by storing a web-based article [8] in Zotero and generating a map using Zotero Maps. This capability therefore allows a non-geospatially trained individual to rapidly extract and visualize geospatial information buried in unstructured text in significantly less time than traditional GIS methods of generating geospatial data.

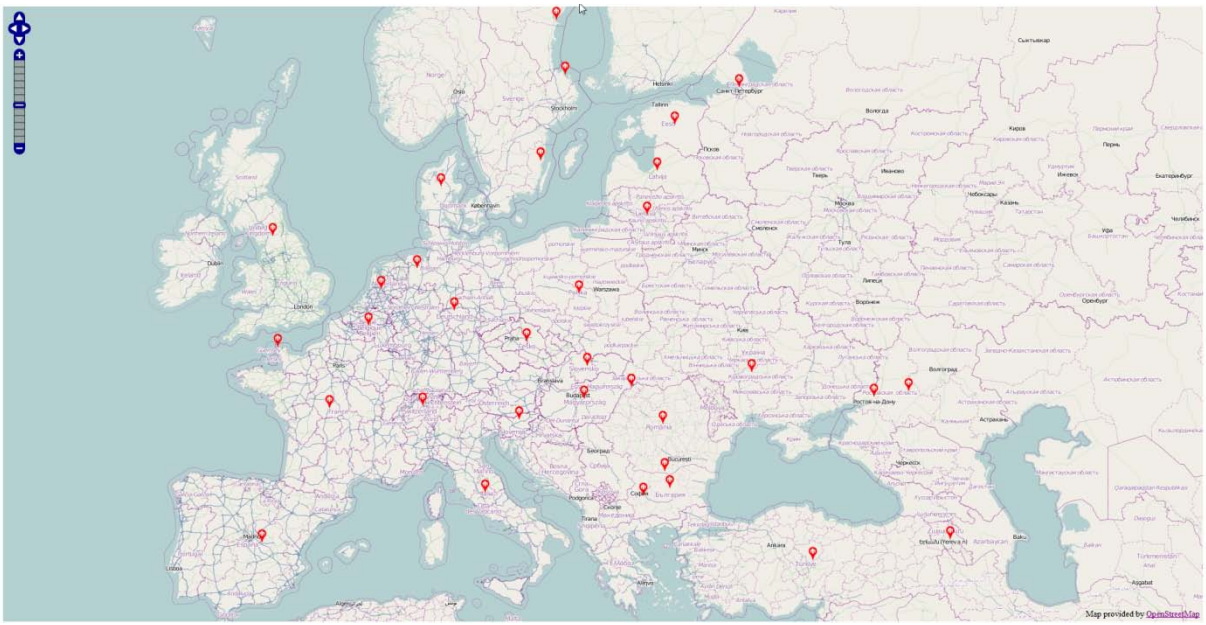

Figure 1 Map generated with Zotero Maps from the web-based article *Plans for New Nuclear Reactors Worldwide* [8].

While dozens of tools exist with potential benefit to this work, two open source software projects for handling geospatial data specifically are OpenLayers and GeoServer. OpenLayers is a pure JavaScript library for displaying and manipulating geospatial data within a web browser [9]. With the ability to display published geospatial resources from anywhere on the internet, OpenLayers is an excellent candidate for the front-end interface of a geospatial safeguards tool. GeoServer is a software server used to organize, edit and publish geospatial data [10]. Written in Java, GeoServer is a potential geospatial back-end candidate for storing and sharing geospatial data within an organization. Both OpenLayers and GeoServer are compliant with Open Geospatial Consortium standards [11] and are therefore interoperable with a wide variety of other geospatial software.

2.2 Geospatial data

Traditionally, geospatial data have come in two primary types, each with relatively few but well defined file formats: vector (or geometric) and raster (or image) data types. While this distinction still holds in general, the emergence of collaborative web technologies has supported the rise of dozens of new

ways of encoding geospatial data, as well as a shift in the ways in which geospatial data is produced and conceptualized. Because high quality geospatial data is no longer created and published only by large government agencies, as has generally been the case in the past, traditional methods of geospatial data discovery fail to lead to many of these new data types and formats.

To guide the development of these tools and to determine how such tools might assist in the geospatial data discovery process, three different search strategies have been devised to discover geospatial data in all formats. First, a general Internet search strategy using search engines such as Google, Google Scholar, and Wikipedia leads to the discovery of unstructured geospatial data in text and images. These data require additional computational procedures to transform them into geospatial data types useable in a mapping context. Second, a geographically enabled search strategy using specific geospatial filters such as coordinate pairs, bounding box coordinates, or administrative boundary names, leads to the discovery of geotagged data and geospatial web services. These data are generally unstructured but have associated geospatial metadata. Examples are geotagged images or blog posts. Third, structured geospatial data, such as ESRI shapefiles and GeoTIFF images, are discovered through geospatial data portals and clearing houses. In general these outlets are run by government or not-for-profit agencies.

Note that these search strategies are not mutually exclusive and one search strategy can lead to the discovery of different types of data.

2.3 Test Case: Paks Nuclear Power Plant, Paks, Hungary

To demonstrate the wide variety of open source geospatial referenced information available using this phased search strategy, a theoretical test case was developed based on the need to collect information to help a safeguards and security analyst to understand Paks Nuclear Power Plant (NPP) near Paks, Hungary.

2.3.1 General Internet search

The first search included sites such as Google, Google Scholar and Wikipedia. Over 580,000 results were received on Google by searching for “Paks Nuclear Power Plant”. The first entry returned was that of Paks NPP on Wikipedia. The second site listed was the home page of the power plant.

The Wikipedia site for “Paks Nuclear Power Plant” had a wide variety of geographically referenced data [12]:

- A location Map
- Latitude and longitude coordinates
- Multiple current and historic images of the site
- Links to the Paks NPP website and many other related sites
- Links to other papers and references

The Paks NPP home page [13] in English also provided some geospatially referenced information:

- An address of the facility
- A location map
- Images of the facility within the “Virtual Tour and Gallery Links” section

From these two websites alone a substantial geospatial reference for the plant can be built. However, note that these data are in text and image formats and thus cannot be easily utilized by traditional geographic information systems (GIS). However, by applying additional computational procedures such as natural language processors to extract place names, these data can be formatted for use in geospatial applications.

2.3.2 Geo-enabled search

From the information gained during the general internet search, specifically the coordinates of the Paks NPP (46.5725N, 18.854167E), Google Earth was used to get an aerial image of the site, dated 20 December 2006. Also available in Google Earth are 3-dimensional building renderings, including a photorealistic rendering of buildings at the Paks plant (Figure 2). Google Earth, which has become the

lay person's geographic information system (GIS) of choice, has the ability to overlay data from dozens of already defined sources including Web Mapping Service (WMS) layers from any external source.

Figure 2: Photorealistic 3D rendering of Paks Nuclear Power Plant in Google Earth.

Next, Wikimapia [14], a crowdsourced mapping service that allows users to digitize and annotate geographic features, was examined. Users have digitized buildings and infrastructure at the NPP site, including reactor housings 1 through 4, cooling water input and output systems, switchyard, control room building, visitors center, fire station, meteorological tower, and bus station, among others (Figure 3). This information can be extracted thorough Wikimapia's API in XML, JSON, KML, and binary formats.

The third geo-enabled search was through the GeoHack [15] website. GeoHack, a tool developed by the Wikimedia community's Toolserver project, aggregates mapping services that are capable of displaying georeferenced content from many different sources. By querying a latitude and longitude coordinate pair, GeoHack returns links to various mapping services that display data centered on these coordinates as well as links to other web-based resources related to these coordinates and thus serves as a valuable jumping off point to a large amount of geospatial data. From here, a large number of other websites containing geo-tagged information were discovered to include:

- 28 global map services sites (Google Maps, Wikimapia, OpenStreetMap, etc)
- 12 Wikipedia links
- 10 photo hosting websites
- 19 "other sites"
- Over 100 regional map services

While each of these sites do not necessarily represent unique data points, as some links are coincident or contain identical data, this does illustrate the relative ease with which recent aerial and satellite imagery and geographic data visualizations are obtainable.

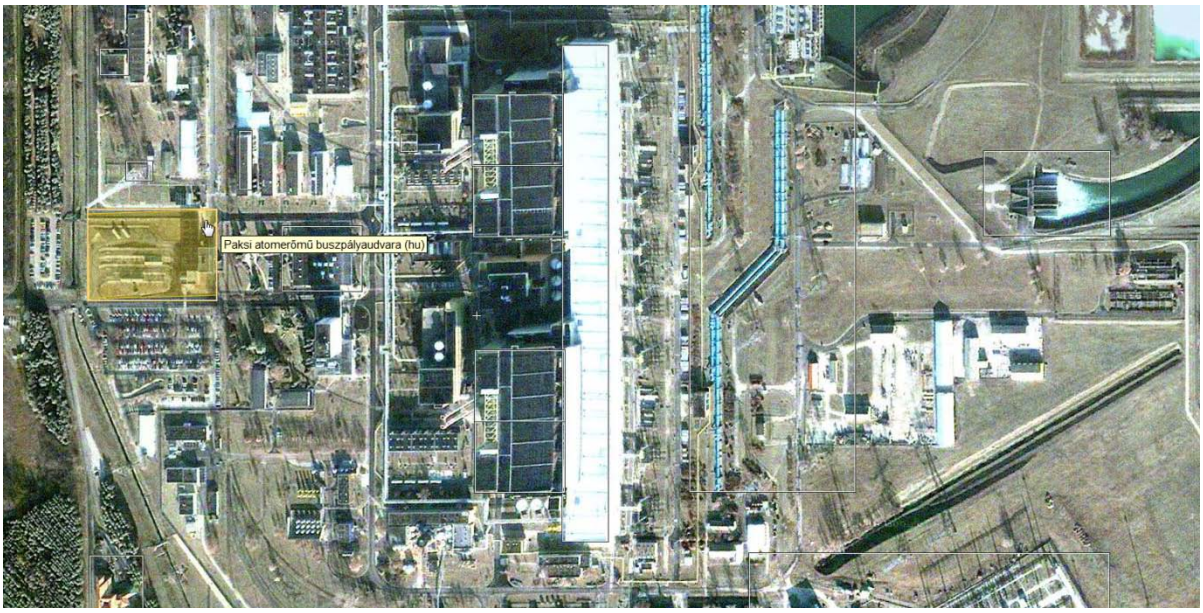

Figure 3: User digitized and annotated features of Paks NPP on Wikimapia.

2.3.3 Structured geospatial data search

Finally, an on-line search was conducted for standard structured geospatial data such as ESRI Shapefiles, digital elevation models (DEM), and GeoTIFF images. Effective use of data in most of these formats requires specialized GIS software (for example, ArcGIS or MapInfo) and a trained geospatial specialist. However, several XML-based geodata formats (for example, KML and GML) have emerged in recent years that allow these data to be used within a web-based computing framework and thus available to a larger number of analysts.

The quality and resolution of the GIS data discovered for the Paks NPP site ranged from very little to extremely high. While a large amount of data were discovered at state and regional scales, very little data were found at local and site-specific scales. For example, geospatial data for Hungary and Hungarian counties were abundant, while data for municipal scales and the Paks NPP site in particular were more difficult to come by. However, what one might consider “micro-level” geodata, such as geotagged photographs, were widely available. This trend might indicate the need to and benefit of examining other sources of geographically referenced data to supplement this mid-scale data void.

2.3.4 Results

Based on discussions with analysts, desirable geospatial information for safeguards and security analysis includes:

- Overhead aerial or satellite imagery
- Reference maps and images to provide context
- Reference information such as roads and other nearby geographic features
- Ground-based photographs
- Detailed site information
- GIS/map data to use in analysis

Each of these data types were discovered on the open Internet with relative ease. However, effective use of these data for analysis requires specialized training and expensive software tools that may not be widely available to analysts. Moreover, no tools exist (to the knowledge of the authors) that allow for the systematic detection, extraction and utilization of these data within a system that can be easily incorporated into the analysis workflow. Also, notably missing from this list are unstructured data (such as text data) containing geospatial references. Because these data are not easily used in a geospatial framework they are often ignored or overlooked. Future work will seek to enable analysts to detect,

extract, and utilize these data for use within a geospatial system in addition to the data types noted above by developing tools that are accessible to analysts and that integrate into workflows with minimal divergence from proven methods.

3 Ontology development

In order to systematically discover and integrate heterogeneous and unstructured data it is necessary to develop and apply a standardized definition of terms and relationships. Ontologies, formal specifications of terms and their relationships within a given knowledge domain [16], allow for the standardization of heterogeneous and unstructured data by defining the spatial, temporal, and thematic dimensions of the data. By applying computer-readable metadata based on these ontologies (semantic mark-up), it is possible to further automate detection and processing of these data by allowing computational reasoning about the geospatial and thematic relationships among data.

For this project, several ontologies are likely needed. First, a geospatial ontology will be necessary to define types of geospatial entities (for example, administrative districts, natural features, etc.) as well as geospatial relationships (for example, adjacency, proximity, containment, etc.). Second, a place names ontology will be necessary to standardize the identification of named geographic entities. Several existing place name ontologies exist in the form of online gazetteers, including GeoNames [17] and Yahoo! GeoPlanet [18]. Finally, thematic or domain ontologies are needed to define safeguards relevant terms and relationships. Examples might include ontologies for the nuclear fuel cycle and nuclear reactor or centrifuge facility operations. Domain ontologies are often specified using Web Ontology Language (OWL) definitions [19].

Existing implementations of the semantic web, which seeks to supplement web resources with computer readable metadata, can support these functions, though the adaptation of these standards has only occurred within relatively narrow parts of the Internet. The utilization of ontologies to increase the effectiveness of safeguards activities has been explored [20], although not within a geospatial context.

4 Conclusions

Using a phased search strategy that includes a general Internet search, geospatially-enabled search, and structured GIS data search, it is possible to assemble a basic geographically referenced set of data without a specialized GIS analyst or expensive GIS software.

Because of the ease of use and low life-cycle costs, the use of these open source tools to create a basic geospatially referenced data set has the potential to increase the use of geospatially referenced data in future safeguards analysis. When configured to work within an existing safeguards analysis workflow, these tools can allow analysts to efficiently and effectively utilize both structured and unstructured geospatial data from the open Internet, a capability that generally is available only to those with specialized training and expensive, proprietary tools.

Several no-cost, open source tools exist that lend themselves to further development for use in safeguards specific analysis. Zotero, an open source reference management tool, for example, has the ability to produce maps from place names in web-based documents or collection of documents in seconds rather than the hours or days when compared to traditional GIS tools, all the while extracting, storing, and organizing references in a structured manner. OpenLayers and GeoServer are open source projects that allow for the handling, visualization, and sharing of geospatial data.

In order for the efficient use of this heterogeneous and unstructured geographically referenced material, more work needs to be done to create standardized and automated processes for discovering, integrating and organizing the data. Development and application of ontologies and semantic technologies will be necessary to achieve this goal.

Finally, while open-source data can be an important supply of new types of information for safeguards analysis, it must be approached with some caution. Open-source data, especially crowdsourced

information, can be inaccurate, incomplete, biased or even fabricated [21]. A future goal of this research is to develop tools and methodologies that provide safeguards analysts the ability to differentiate valid and reliable geospatial data from those data that cannot be trusted.

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22 Integrated Measurement and Monitoring Systems

DEVELOPING A SYSTEMS OPTIMIZATION TOOL FOR MONITORING SPECIAL NUCLEAR MATERIAL

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ABSTRACT

Under its international safeguards obligations, a nuclear facility will implement a security system that is developed and designed to incorporate (and hopefully integrate) elements of physical protection and containment and surveillance. Simple nuclear security components can range from closed-circuit camera systems, electromagnetic door locks, motion sensors, physical barriers, portal monitors and other radiation sensors, and radio-frequency identification tags. Integrating these various components into an effective system is difficult yet essential in providing confidence in the security and control of the special nuclear material within the facility. A tool for optimizing various material control and containment and surveillance systems would facilitate effective implementation of these systems for high assurance that material diversion could occur. At Texas A&M University, staff at the Nuclear Security Science and Policy Institute (NSSPI) has begun investigating such a systems optimization tool for various material control and containment/surveillance systems that is to be implemented in a small static, storage facility: an applied safeguards teaching laboratory for graduate-level nuclear engineering students. The facility simulates a typical static professional research laboratory with special nuclear material. The tool is based on using a stochastic radiation transport code for determining vulnerabilities of the installed radiation monitoring systems within the laboratory. In early 2010, NSSPI staff completed a proof of concept by simulating the movement of one highly-enriched uranium source though and out a single room with a single point of exit. The results were indicative of suspected vulnerabilities by the investigators and a more complex design and scenario was then devised for the next scenario: increased radiation attenuation, elevated radiation backgrounds, accelerated motion, more points of access, etc. This presentation will discuss the results of this advanced modeling endeavor and present the work into a hypothetical systems optimization tool that could eventually benefit the nuclear safeguards and security industry.

KEYWORDS: Nuclear Material Monitoring; Systems Optimization

1. Introduction

The Nuclear Security Science and Policy Institute (NSSPI) at Texas A&M University (TAMU) currently operates an applied safeguards technologies laboratory where various material control and accounting (MC&A), physical protection (PP), and containment and surveillance (C/S) systems are used for educational and research purposes for graduate students receiving nuclear engineering degrees yet specializing in nuclear nonproliferation, safeguards, and security. This laboratory is to be used for educating students in various technologies needed to effectively apply nuclear security and safeguards measures via practical exercises using all the equipment and special nuclear material (SNM) housed in the lab space. A basic C/S system has been put in place to secure a small number of SNM sources within the lab and a basic vulnerability assessment of the system has been conducted. The information from a vulnerability assessment is used to optimize the C/S system for maximized material diversion interdiction. The theory behind this system optimization will eventually be translated to other technologies and systems within the laboratory.

2. Vulnerability Assessment on the TAMU ASTL

In the nuclear security industry, vulnerability assessments are used mainly to detect and identify weaknesses in a physical protection system. The challenge at TAMU was to apply this logic to a C/S radiation measurement system within the Applied Safeguards Technologies Laboratory (ASTL). Apart from the primary objective of identifying and evaluating these weaknesses, a secondary result of a vulnerability assessment includes optimization to mitigate the system's vulnerabilities. This step is discussed within the results section of this paper.

As aforementioned, the ASTL houses portal radiation monitors, SNM standards, four gamma monitoring systems, cameras, remotely-controlled MC&A devices, and physical protection instruments such as balanced magnetic switches and other access restricting devices. The primary intent is to use this lab mainly as a teaching laboratory for holding semester-long courses on applied advanced safeguards and security technologies for graduate-level students within the nuclear engineering department at TAMU. Additionally, the laboratory is available for students in need of using the SNM or equipment for their graduate research projects in safeguards or security.

3. Simulation

The laboratory was modeled in three dimensions using a stochastic radiation transport code called Monte Carlo N- Particle (MCNP) developed by staff scientists at Los Alamos National Laboratory[1]. The (MCNP) code allows for structural and radiation modeling and simulation is used to determine the effectiveness of the radiation detection system in place within the TAMU ASTL. For simplicity concerns, the laboratory was modeled as a 9.23-meter (30 feet) by 7.385-meter (24 feet) concrete room with 3.69-meter (12-foot) high concrete ceilings. There is a single point of entry and no other external access points (i.e., windows or gates). The furniture fixtures were kept at a minimum including a wooden work bench along a freestanding concrete wall outlining the entrance/exit ramp into the lab and the steel safe where the sources are housed. Figure 1 shows a bird's-eye view of the laboratory with approximate dimensions (not to scale).

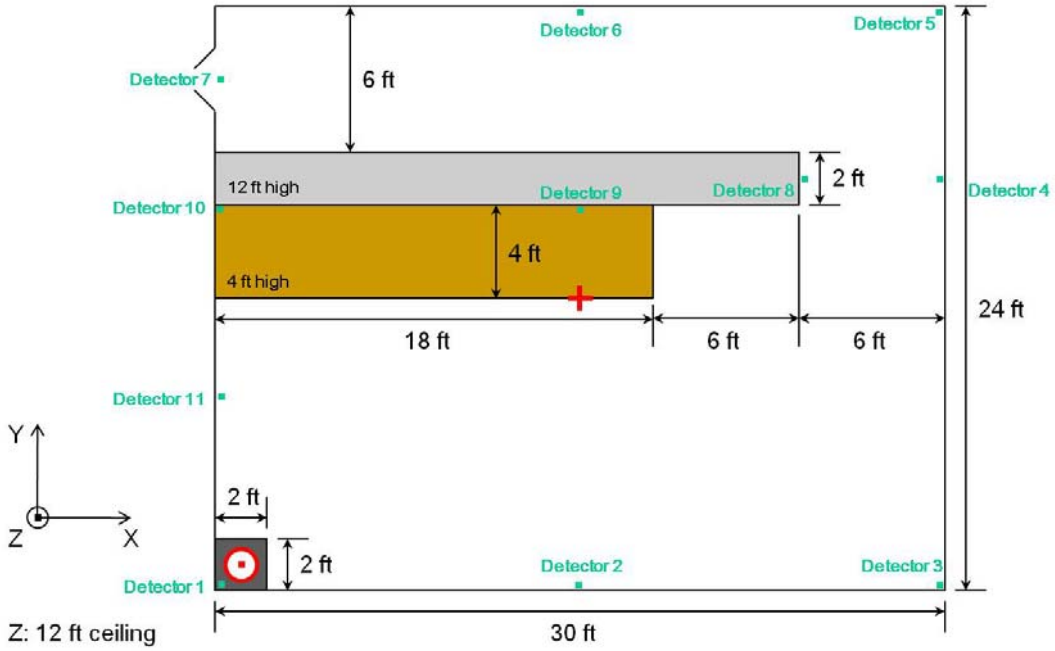

Figure 1. TAMU ASTL with source in safe (the red/white target is the source)

The radiation detection system consists of Canberra G64 Monitors (Geiger-Muller (GM) tube-based detectors) used for gross photon counts and intends to detect any movement of a given radioactive source via the placement of the detectors. A total of eleven point detectors were simulated in the model in order to choose an optimal subset/location of detectors for the given laboratory space. Enriched ^{235}U- radiation source model based on the 4.46%-enriched uranium standard from the NBL

CRM 969 set was considered for the simulation placed at various locations along a suspected adversarial pathway within the lab. The results are given per single photon emitted per second and furthermore, the detector tallies are assumed as flux tallies in units of photons/cm^2-s. The adversarial pathway begins from the source kept in the storage safe and twenty source location points outside the safe as the source nears the point of exit. Figure 1 shows the starting point of the simulation (the source is the red/white target within the steel source storage safe in the lower left-hand corner). Figure 2 displays the pathway (21 source positions outside of the safe) with the eleven hypothetical detectors.

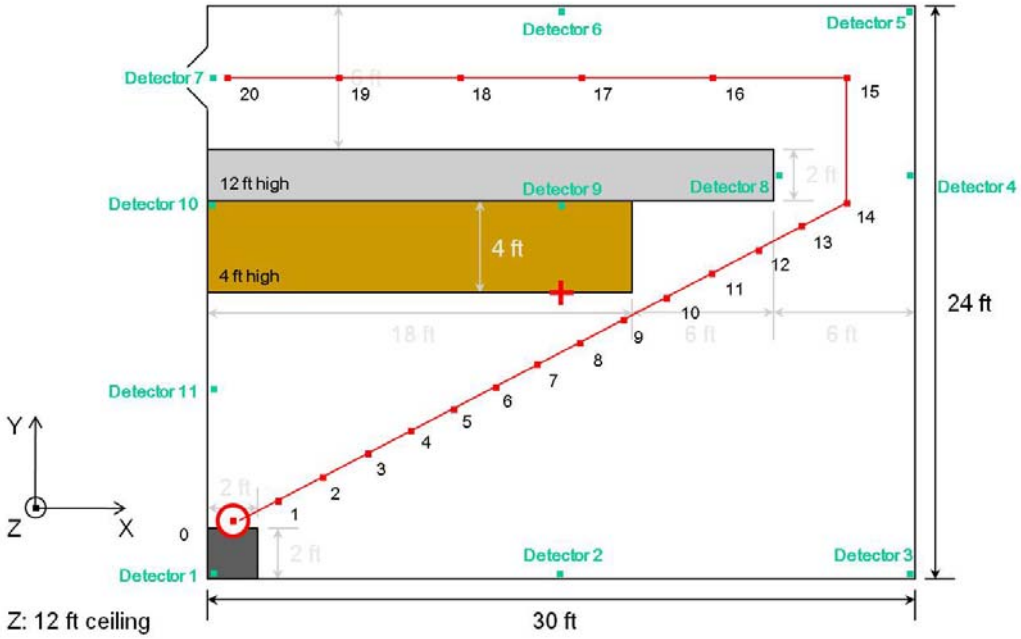

Figure 2. TAMU ASTL with defined source pathway (source is outside safe)

Initially, the defined pathway used is simple straight-line pathway to the point of exit. For this activity, it was assumed that the simplest case would be evaluated for determining the feasibility of the theory and further scenarios would eventually be implemented if the initial case provided adequate results. With this path, from the eleven detectors flux results were tallied and then a subset from it was chosen to optimize the detector system.

4. Results

The simulation results shown in figure 3 are in units of photons/cm^2-sec per source strength of one photon per second. There are twenty-one positions. Conceptually, the data supports the theory of tracking material movement via the detector system. With a given threshold of 1E-6 photons/cm^2-sec per one photon per second, figure 3 conveys that an aggregate of detector locations 11, 9, 4, and 6 will create the closest semblance of continuous material tracking using the minimal number of detectors from initially removing the source from the safe to removing the source entirely from the room.

Figure 4 shows the resulting detector profiles when the superfluous detectors are removed. It is important to note that at source location 20, the reading from Detector 6 drops below the aforementioned threshold (circled in red). Ideally, it would benefit the facility to utilize Detector 7's location but within the limitation of resources posed by the exercise, it is beneficial to show the efficacy of optimizing the placement of Detector 6 to meet the needs of the system.

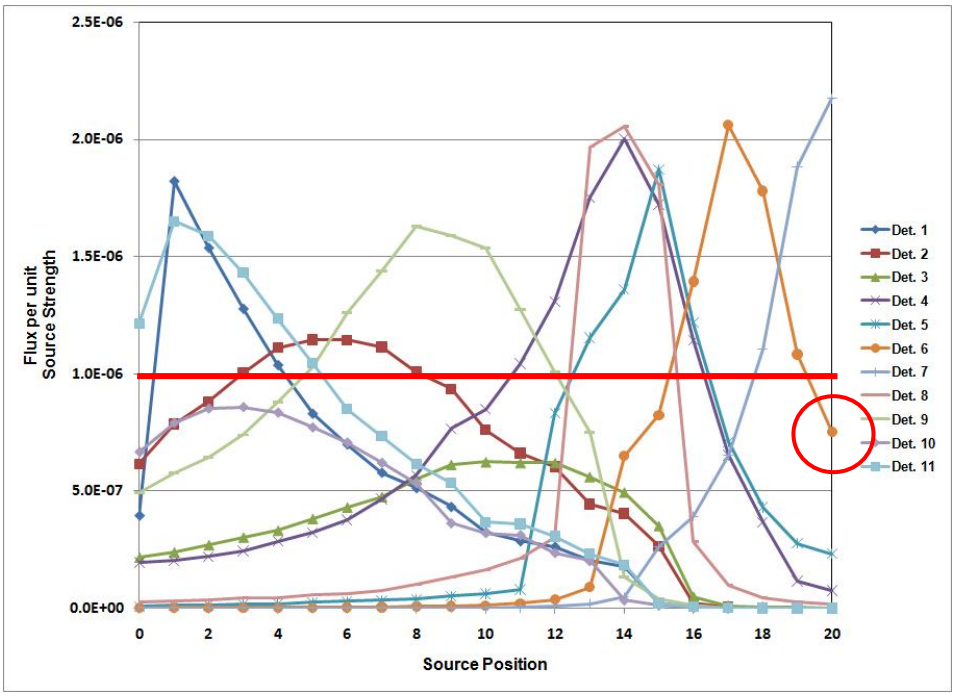

Figure 3. Flux per unit Source Strength vs Source Position for 11 detectors

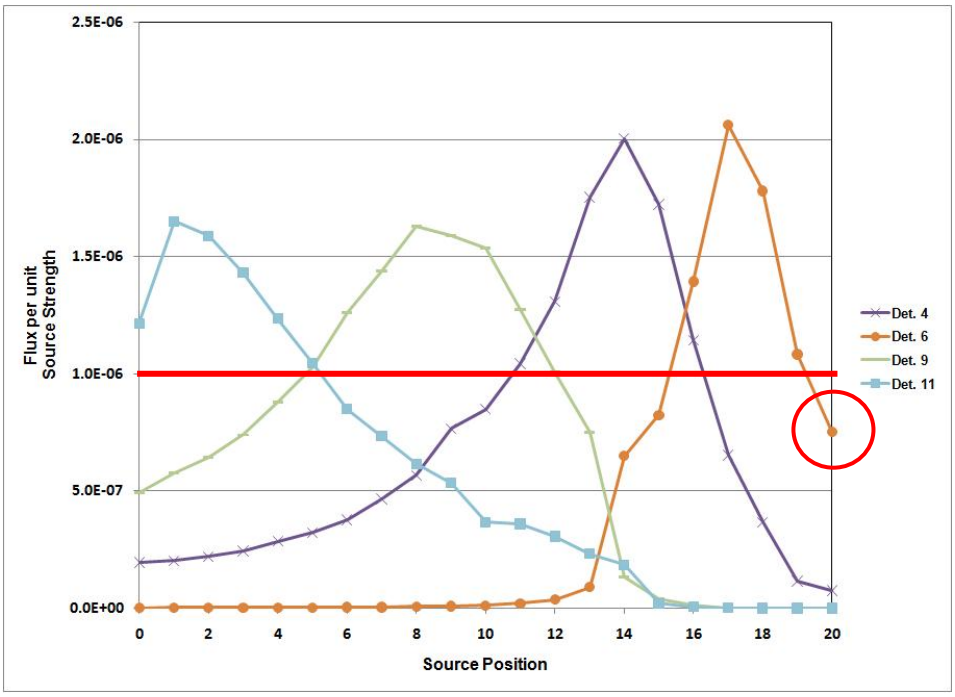

Figure 4. Flux per unit Source Strength vs Source Position for Detectors 4, 6, 9, and 11

Within the confines of only using four detectors, Detector 6 was repositioned to an optimized location (80 centimeters in the –X direction) that will mitigate the low count rates exhibited by source location 20. Figure 5 shows the repositioning of Detector 6 and figure 6 shows the effect of this on the profile of the source tracking.

Overall, the results convey that the exercise was a success and that material, albeit in a rudimentary setting, can be tracked through a given, simple facility based on these simulations.

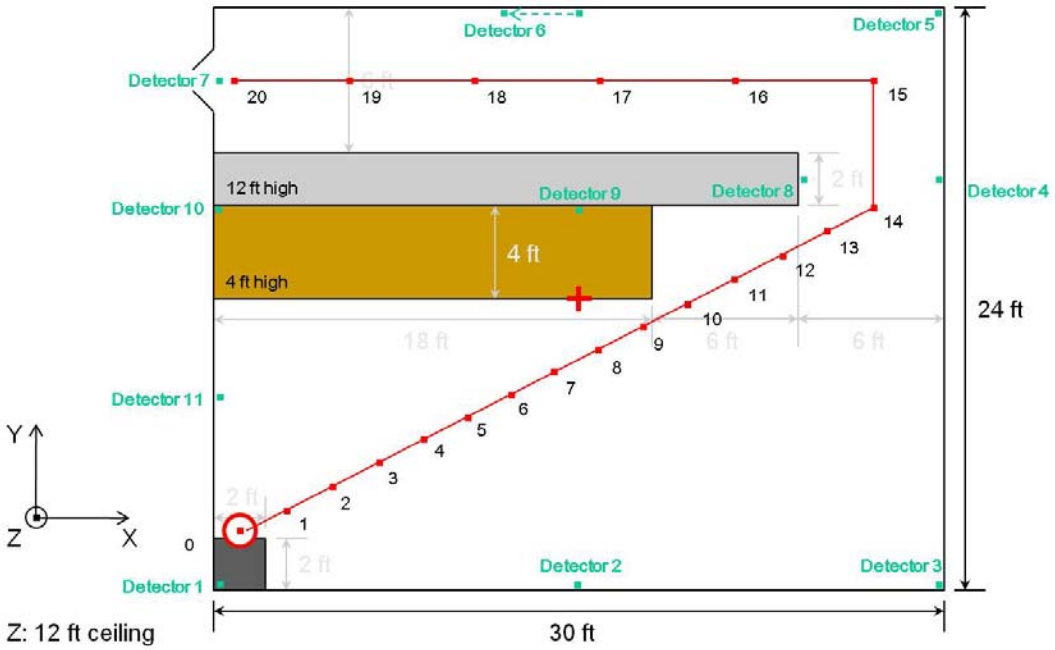

Figure 5. Detector 6 repositioned for 4-detector optimized system

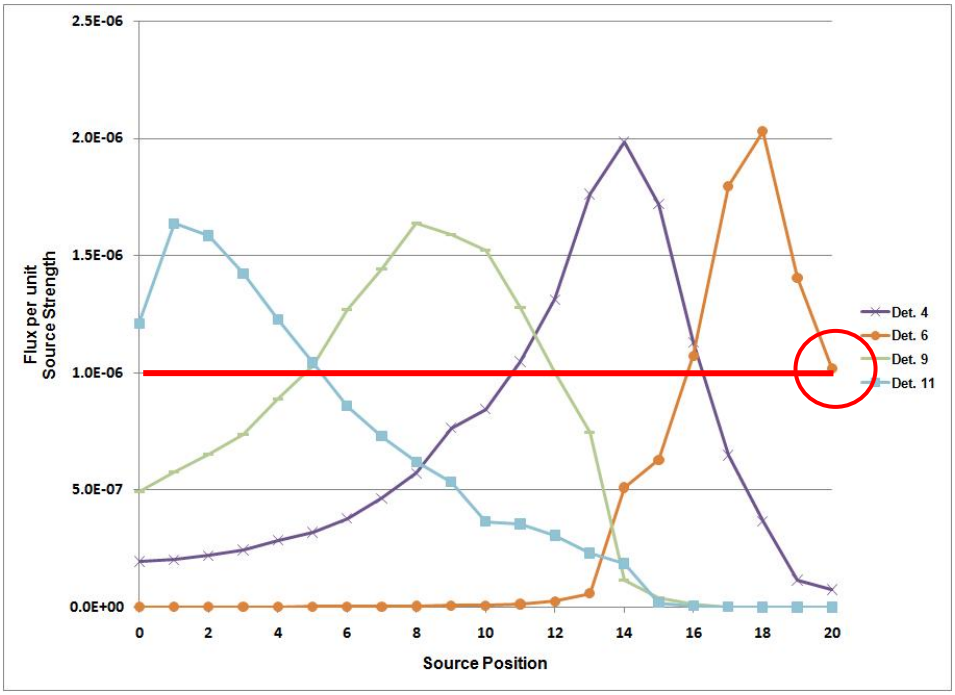

Figure 6. Flux per unit Source Strength vs Source Position for Detectors 4, 6 (modified), 9, and 11

5. Future Work

The investigators made assumptions in the simulated exercise to simplify the intricacies of the ASTL at TAMU and the sources that are to be used eventually in a system such as this. In future incarnations of the system, more detailed detectors, sources, and facility space will be incorporated to better fit the needs of the system. In light of the simplification measures taken by the team, it is believed that the resulting data convey favorable results for optimizing a material tracking system using a stochastic method/code such as MCNP5. Ultimately, as this project evolves, added considerations will be made to factor sensitivities of the detector configurations and details of the radioactive sources. Further, it is envisioned that the research team will utilize knowledge gleaned from this exercise more accurately model the laboratory space and determine the system's

vulnerabilities. Important among the considerations when continuing with this work in the future include modifying the detector tally points in MCNP to fully modeled scintillator detectors; characterizing the performance of the system with considerable amounts of background radiation (apply realistic scenarios instead of a low-background experimental situation); and including other, less detectable, sources beyond the 1.001 MeV photon typical of ^{238}U. As previously mentioned, the researchers will also use experience taken from this exercise and apply it to other relevant technology as applied in nuclear material tracking in a given static facility monitoring such as optical surveillance. It is believed that lessons learned from the basic stochastic modeling methods used in a Monte Carlo code, can be applied to other fields and this exercise is a clear first step in that direction.

6. Conclusions

In conclusion, the results of this exercise exhibit that a stochastic, Monte Carlo code (such as MCNP) can be used as a valuable system optimization tool for a basic C/S or material tracking system using radiation monitors in a static facility. Lessons from this work can be used in safeguarding any nuclear facility where optimization of a limited number of radiation detectors is essential. This limitation is a real-world situation that almost all facilities face continuously in today's world. Whereas the initial reaction for enhancing an existing system would be to merely add detectors, this exercise determined that detector placement is just as important to the adequate and effective operation of a radiation C/S system in a given facility. Furthermore, an optimized radiation detection system can be supported by a secondary system (e.g., optical) that would benefit greatly from the applied data gleaned from this exercise. Overall, this exercise showed the feasibility of optimizing a material tracking system by optimizing a system of detectors using the radiation transport code MCNP. In TAMU's nuclear engineering MS degree program in Nonproliferation, this activity has been incorporated into an advanced safeguards course to educate graduate students on the value of optimizing a system using a code with which they are highly familiar.

7. Acknowledgements

The research team would like to extend gratitude to Dr. William S. Charlton of Texas A&M University and the Nuclear Security Science and Policy Institute for providing the means and resources in conducting this work as well as provided the initial idea for this exercise.

8. Legal Matters

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9. References

1. X-5 Monte Carlo Team, 2005 MCNP-A general purpose Monte Carlo N-Particle Transport Code, Version 5, LANL Report No. LA-UR-03-1987.

Development of Solution Monitoring Software for enhanced safeguards at a large scale reprocessing facility

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Abstract:

The implementation of an effective and efficient IAEA safeguards approach at large scale reprocessing facilities with large throughput and continuous flow of nuclear material requires the introduction of enhanced safeguards measures to provide added assurance about the absence of diversion of nuclear material and confirmation that the facility is operated as declared. One of the enhanced safeguards measures, a Solution Monitoring and Measurement System with associated advanced software, is being implemented at a large scale reprocessing plant in Japan. The Solution Monitoring Software (SMS) is designed as a tool to enable automatic calculations of volumes, densities and flow-rates in selected process vessels, including most of the vessels of the main nuclear material stream. This software also includes automatic features to support the inspectorate in verifying inventories and inventory changes. The software also enables one to analyze the flows of nuclear material within the process and of specified "cycles" of operation, and, in order to provide assurance that the facility is being operated as declared to compare these with those expected (reference signatures). The configuration and parameterization work (especially the analytical and comparative work) for the implementation and configuration of the SMS has been carried out jointly between the IAEA, Euriware-France (the software developer) and the Joint Research Centre (JRC)-Ispra. This paper describes the main features of the SMS, including the principles underlying the automatic analysis functionalities. It then focuses on the collaborative work performed by the JRC-Ispra, Euriware and the IAEA for the parameterization of the software (vessels and cycles of operation), including the current status and the future challenges.

Keywords: solution; monitoring; software; reprocessing

1. Introduction

The implementation of an effective and efficient IAEA safeguards approach at large scale reprocessing facilities with large throughput and continuous flow of nuclear material requires the introduction of enhanced safeguards measures to provide added assurance about the absence of diversion of nuclear material and confirmation that the facility is operated as declared. One of the enhanced safeguards measures, a Solution Measurement and Monitoring System (SMMS) with associated enhanced software, is being implemented at a large-scale reprocessing plant in Japan.

The SMMS is applied on the chemical liquid processing part of the plant operation and involves over 90 vessels or other equipment which can, at any given time, contain over 95% of the Pu inventory of the main process (liquid) material balance area. The installed measurement instruments provide signals for pressure, temperature and/or neutron count rates.

Two different types of solution monitoring instruments have been installed in the reprocessing plant. The first type; known as SMM1 instruments; are high accuracy IAEA owned differential manometers which are connected directly to the Operator's pneumatic dip tubes. These instruments are applied on the most strategic vessels in the main process line. A robust data collection system is connected to automatically provide data to a data base. In regard to the implementation of the SMM1 system, major consideration was given to the data collection redundancy and integrity. The second type of instruments, known as SMM2 type instruments, is owned by the Operator. The operator's current analogue output signals are split and provided to the inspectorate cabinets. In this case, authentication measures have to be applied by the Agency to the outputs of these instruments. More detailed information on the architecture of the SMMS was provided by Ehinger [1]. The associated software, referred to as Solution Monitoring Software (SMS), automatically processes and evaluates SMMS data to support the inspection activities in such a large-scale reprocessing plant.

2. Solution Monitoring Software

2.1. Overview

The software for the analysis of the SMMS data is specifically designed to handle large amounts of data and to support its review by IAEA inspectors so that the effort required to draw safeguards conclusions is reduced. The SMS is a part of the Integrated Inspector Information System, which is the collection of software modules providing automated support for the inspection activities. The requirements for the information technology architecture for solution monitoring are discussed by Thevenon [2].

The SMS computes density, volume and flow rates whereby one of the features of the software is to provide support for the safeguards verification activities (inventory verification, inventory changes and flows within the process). The advanced SMS features enable evaluation of operational cycles against expected ones in order to confirm that the facility is operated as declared and provides additional assurance about the absence of diversion of nuclear material.

2.2. SMS Modules

The software comprises the following modules: configuration, pre-processing and calculation, and evaluation. The configuration module is part of the user Interface whereas an automated evaluation function is implemented in order to support data reviews by the inspectors.

The pre-processing and calculation module extracts time-stamped raw data (e.g.: pressures, temperatures) at determined time intervals and transfers it to the time series tables of the IAEA data base. Derived quantities such as volume and density are then calculated further from the raw data.

The evaluation module provides a diagnosis which must facilitate the task for inspector reviews and for drawing conclusions. The analysis method for the SMS data evaluation at the facility, and the algorithm capabilities in "behaviors" recognition and flags of discrepancy were discussed in detail elsewhere [2, 3]. Within the SMS design, the data evaluation is based on the identification of the declared operating cycles of the equipment (detection of the start of a filling from a certain vessel, end of a transfer, start of a transfer into another vessel, detection of the end of an operational cycle). This is called the auto-correlation function of the SMS evaluation module, which checks that the sequence of functional behaviour (events) respects a predefined design. A notification is given to the inspector in the case of an out-of-sequence event and also in case of the successful completion of a cycle. Solution transfers between certain vessels (sending and receiving vessels) are identified and are also checked for volume/mass consistency against predefined tolerances by the evaluation software (cross-correlation function).

The configuration module enables the set-up of the calculation and evaluation modules. Vessel specific information like calibration data, probe separations and outbound values for density calculations are input first. Also, for each piece of equipment the partner vessels which are potentially sending/receiving solution are identified. At a next level, the parameters for the evaluation modules are set-up by using a specific stand alone application provided by the software. This aspect will be discussed in more detail in section 2.4.

2.3. SMS Inspector Interface- Inspector Log

Due to the large amount of data to be evaluated, the SMS is designed with a drill down capability from higher level data structures to the raw data. The functionalities of the Inspector Interface for data evaluation are presented as follows.

The first level of the Inspector Log summarizes the solution status in the monitored vessels within a defined time period (Fig. 1). In order to review data for a certain vessel, the inspector can access the Data Graph display for the concerned vessel (Fig. 2) by selecting the “Status”. From the Data Graph page the derived quantities and raw data can be further accessed in the box on the right side of the graph, as illustrated in Fig. 2.

Date and time	Cycles and transfers	Status
2008-04-15 11:16:00	Vessel X 59 (l)	OK
2008-04-15 11:01:30	Vessel Y 47 (l)	OK
2008-04-15 09:36:30	Vessel Z Completion of measurement cycle	OK
2008-04-15 09:36:30	Vessel Z 947 (l)	OK
2008-04-15 09:36:30	5,057 (l)	OK
2008-04-15 09:19:30	875 (l)	OK
2008-04-15 08:01:30	11,960 (l)	OK
2008-04-15 08:01:30	Vessel A to B 11,905 (l) 11,860 (l)	OK
2008-04-15 07:55:28	Completion of measurement cycle	OK
2008-04-15 07:55:28	13 (l)	OK
2008-04-15 07:39:00	Completion of measurement cycle	OK
2008-04-15 07:32:30	1,487 (l)	OK
2008-04-15 07:11:30	99 (l)	OK
2008-04-15 07:07:00	11,919 (l)	OK
2008-04-15 06:36:59	622 (l)	OK
2008-04-15 06:07:00	6,098 (l)	OK
2008-04-15 05:20:33	219 (l)	OK
2008-04-15 05:15:00	0 (l)	OK
2008-04-15 04:16:30	218 (l)	OK
2008-04-15 04:12:29	193 (l)	OK

Fig. 1: Results of the automatic evaluation of cycles, solution transfers and solution status

Results of the automatic evaluation process events are colour coded, enabling the Inspector to focus on possible discrepancies. By using the Inspector bars and available buttons, the automatically created events can be consulted, accepted or can be corrected, in case of a misplaced event. For instance, Fig. 3 shows a cross-correlation alarm indicating an inconsistency in the calculated mass of the transferred solution determined by an incorrect positioning of the transfer event. Following a repositioning of the event by the inspector, the transferred volume/mass is automatically re-calculated by the software.

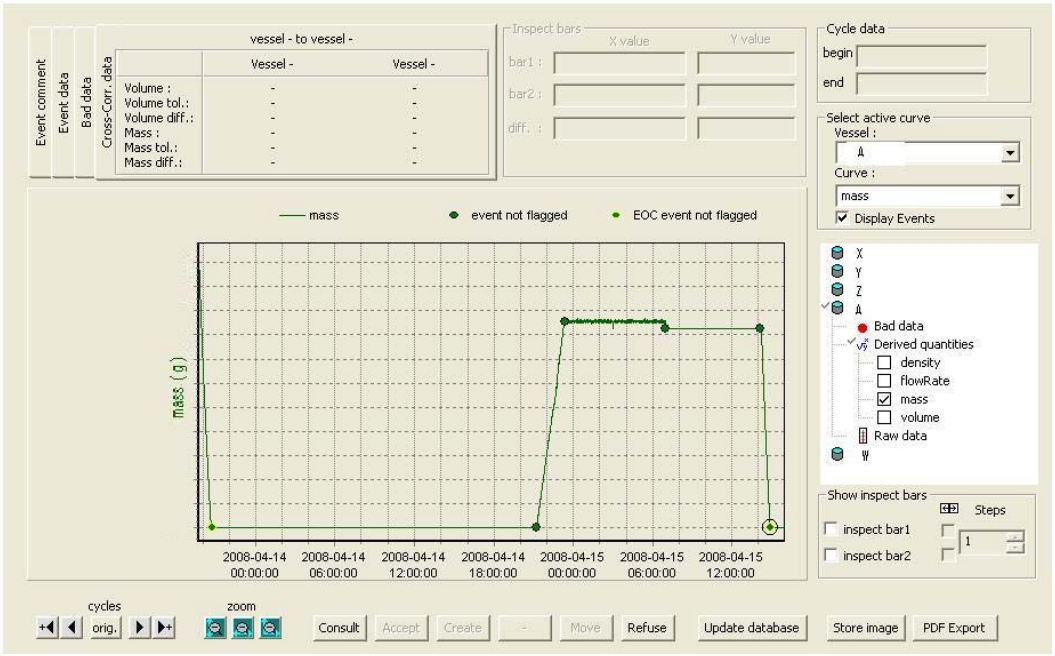

Fig. 2: Data Graph display of accountability tank cycle–automatically positioned events (green circle) associated with the detection of functional behaviour

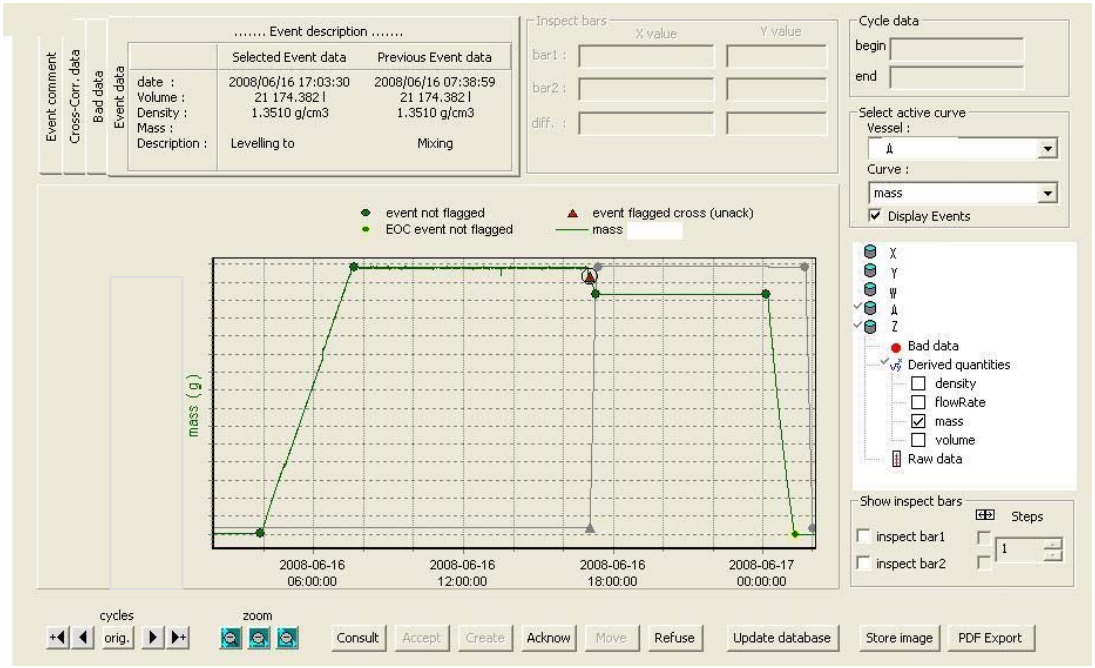

Fig. 3: Data display of an accountability tank cycle: a solution transfer is flagged as a cross-correlation alarm

The inspector has the possibility to review every detected transfer which took place within a certain time period between defined partner vessels, as shown in Fig 4.

Date	Sending Equipment			Receiving Equipment		
Begin Date - End Date	Equipment	Volume (m3)	Sol. Mass (kg)	Equipment	Volume (m3)	Sol. Mass (kg)
2008-04-13 12:32:00 - 2008-04-13 13:43:30		19.69	27,255.79		19.66	27,253.62
2008-04-13 11:45:30 - 2008-04-13 12:08:30		1.66	2,291.83		1.67	2,309.15
2008-04-13 07:47:00 - 2008-04-13 09:22:30		4.96	6,891.06		5.00	6,901.22
2008-04-13 01:28:30 - 2008-04-13 02:20:30		2.82	3,927.25		2.83	3,908.73
2008-04-12 23:14:30 - 2008-04-13 03:04:00		21.33	29,606.57		6.33	10,890.14
2008-04-12 23:13:29 - 2008-04-13 02:57:30		21.33	29,606.57		15.00	18,651.03
2008-04-12 14:19:00 - 2008-04-12 16:04:30		5.96	8,288.34		5.97	8,240.99
2008-04-12 11:10:29 - 2008-04-12 11:33:00		0.95	1,473.74		0.95	1,468.55
2008-04-12 04:50:00 - 2008-04-12 08:13:30		0.98	1,096.91		0.97	1,084.86
2008-04-12 03:10:00 - 2008-04-12 04:49:00		5.85	8,156.90		5.88	8,121.08
2008-04-12 01:50:59 - 2008-04-12 02:06:59		0.67	1,033.03		0.67	1,025.95
2008-04-11 22:02:00 - 2008-04-12 01:24:30		19.59	27,254.53		19.77	27,269.72
2008-04-11 21:21:30 - 2008-04-11 21:35:00		0.10	108.17		0.09	126.21
2008-04-11 17:12:30 - 2008-04-11 18:50:30		5.16	7,205.57		5.17	7,154.98

Fig. 4: Historical Summary of solution transfers between defined partner vessels within a defined time period

2.4. Configuration and parameter setting for the evaluation module

The SMS software has been developed under contract with Euriware, France and with support from the European Commission-Joint Research Centre (JRC) in Ispra, Italy. The collaborative work not only envisages taking advantage of the existing expertise in solution monitoring developed at the JRC-Ispra, but also of the knowledge on the operational characteristics of the reprocessing facility.

In view of the configuration and parameters setting (parameterization), vessels have been prioritized according on their safeguards significance. The first step of development is the configuration, which consists in creating a skeleton of the cycle related to a vessel, by defining all possible successions of functional behaviors (standard events) in order to describe the possible events in regard to the solution in the subject vessel, in operation or not. This work has been completed through several joint meetings between the IAEA, Euriware and the JRC-Ispra. For this purpose, the behaviour of the concerned vessels has been analyzed based on the available relevant operational facility data (acquired during operational periods) and on vessel specifications. All possible transfers of interest from/into a selected number of vessels have been identified. New standard events have been created in order to account for all possible specific process operational parameters (e.g., sampling, acid dilution). On this basis, sequences of standard events (e.g., transfer, mixing, stand-by) were defined to describe the expected behaviour of a cycle (reference signature) of a given vessel. An example for vessel configuration is discussed in the followings. Fig. 5 illustrates an actual operational cycle of an accountability tank.

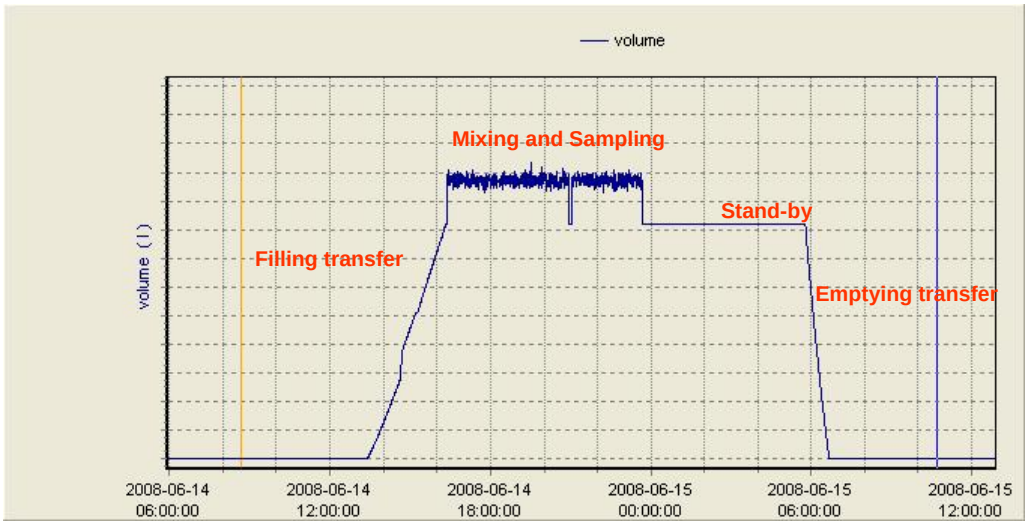

Fig. 5: Data display of a typical cycle for an accountability tank

The reference signature of this tank is represented in Fig. 6. Each functional block represents a standard event and the link between them a possible pattern to be observed. The following events are defined: filling from feeding vessel(s), mixing and sampling, stand-by, emptying transfer into a receiving vessel. The end of the cycle defines the completion of the expected operational cycle for the auto-correlation check.

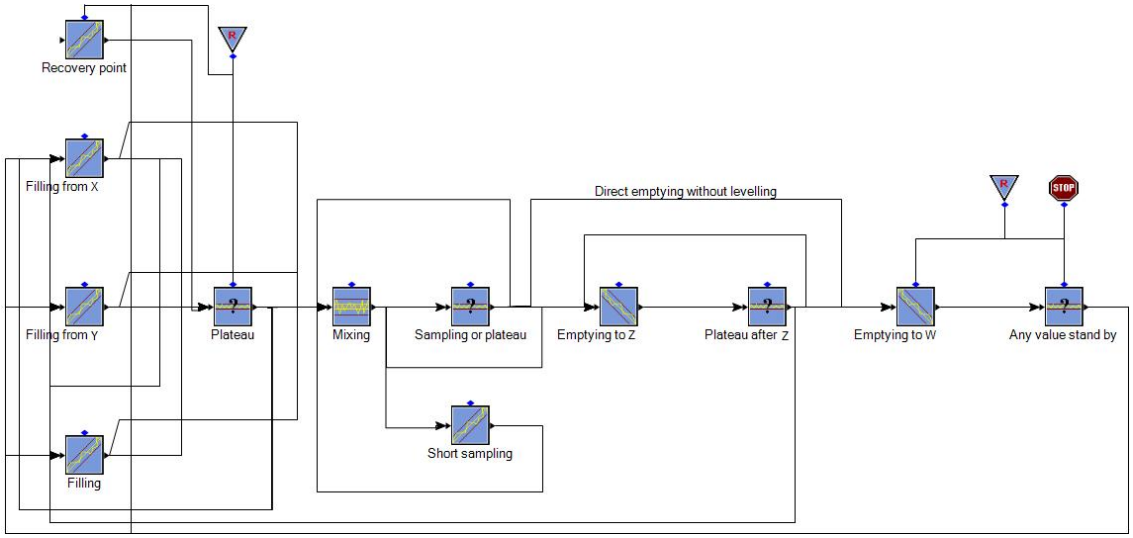

Fig. 6: Reference Signature for an accountability tank

Once the reference signature is fully configured, the attributes of the specific parameters corresponding to each standard event are defined (e.g., the average value of a slope and its tolerance, the tolerance of a stand-by, and the standard deviation for the mixing). Specific parameters can be chosen to better describe the specific operational conditions (e.g., 'Maximum allowable time for being monitored status', 'Duration outside criteria amount', and 'Latency time') and their attributes can be refined later on for a more correct event detection and positioning. Fig. 7 illustrates an example of all parameters that may be used to characterize a single standard event.

Further parameterization steps consist of defining and setting the cross-correlation of interest. Between successive shipping and receiving vessels the solution transfers are confirmed based on the transfer slopes and are verified for consistency by comparing the difference between the transferred masses/volumes of the sending and receiving vessels against defined tolerance limits.

During the parameterization work the aim is to detect cycles corresponding to the declared process events while the number of generated false alarms is kept to a minimum. At the same time, an alarm should be raised in case of an unexpected or out–of-sequence event. The knowledge of possible allowable changes during the operation of the subject equipment which may change the signals is very important in order to obtain a correct evaluation whereby the effort into the investigation of irrelevant alarms is reduced.

Challenges are presented by the multitude of interconnected vessels as some have several feeding/ receiving partners, each with a different particular behaviour including different solution transfer modes. In particular, vessels with continuous input or output and simultaneous batch outputs and inputs are extremely difficult to parametrize. The analytical work is complicated by the fact that even for a given vessel, cycles are not identical: transfers can be delayed or interrupted and particular vessels are characterized by very noisy signals. All these considerations have been taken into account in the parameterization work of individual vessels and prompted, in some cases, the additional creation of specific standard events in order to account for, e.g.: "return from air lift", "acid dilution", "continuous filling while emptying".

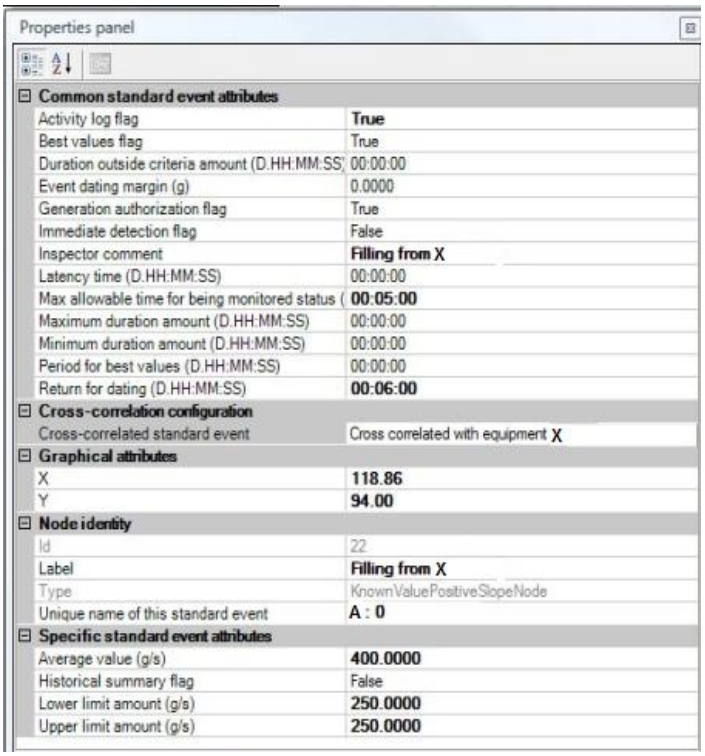

Fig. 7: Properties window of the standard event parameters

As for the parameterization of the reference signatures, Euriware was contracted with the parameterization of a large part of the equipment. For the remainder of the monitored vessels, the parameterization work is carried out jointly with the Process Monitoring Laboratory staff of the JRC in Ispra. The joint IAEA-JRC work also involves testing and tuning of reference signatures developed by Euriware, as well as their integration to the cross-correlation of partner vessels. Beside the progress achieved with respect to the global development of the SMS reference signatures, the collaborative work with the JRC proves to be very valuable for the IAEA as an on the-job-training and a return of experience in respect to solution monitoring. Apart from the work directly related to the configuration and parameterization, efforts are dedicated to the evolution of the software, especially concerning the event detection capabilities. During the tests of the reference signatures software troubleshooting and correction of identified issues have been implemented during the course of the project. While considerable work has been devoted by the JRC in regard to testing and diagnosing software issues and also, in some cases, proposals for corrections, it is Euriware's responsibility to finally investigate these issues and to implement the corrective measures or proposed evolutions, upon acceptance from the IAEA.

Significant progress on vessel configuration has been achieved over the last two years. The reference signatures for most of the vessels involved in the main stream of Pu (18 reference signatures) have been already tested and have been refined following the availability of a new set of data and several software upgrades. Whereas for other vessels (approximately 20) the respective reference signatures were developed and tested, their fine tuning is still pending the availability of a new set of representative operational facility data. There were also some vessels identified for which reference signatures could not be developed due to the lack of representative data.

3. Conclusions

Advanced software was developed for SMMS at a large scale reprocessing facility. The software is designed to handle large amounts of data to enable their automatic processing and evaluation. This

constitutes a valuable tool in supporting the inspectors in the review of data and reduces the effort required in verification activities and the drawing of safeguards conclusions.

The implementation and configuration of the software is being carried-out jointly between the IAEA, Euriware France (the software developer), and the JRC-Ispra. The joint work benefits from the expertise in solution monitoring developed at the JRC-Ispra. Apart from the achievements regarding the development of the reference signatures for the configuration of the software, the collaborative work is also very valuable for the IAEA to gain insights in the field of solution monitoring.

Future work will focus on aspects related to software evolution as well as on testing of the already developed reference signatures, based on the currently available data. However, as far as the fine tuning and development of new reference signatures is concerned, the progress and completion of work relies on the availability of new sets of representative data following regular operational periods of the facility.

4. Acknowledgements

The authors would like to express their gratitude to the several IAEA colleagues who have been supporting the implementation of the SMS software upgrades during the past years. In alphabetical order: A. Alessandrello, C. Dalton, J. Garner (departed from the IAEA), as well as those involved at earlier stages in the SMS project: M. Ehinger (departed from the IAEA) and F. Abazi.

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New Measures to Safeguard Gas Centrifuge Enrichment Plants

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Abstract:

As gas centrifuge enrichment plants (GCEPs) increase in separative work unit (SWU)capacity,new safeguards measures are being investigated to supplement the current International Atomic Energy Agency (IAEA) model safeguards approach to maintain effectiveness, recognizing budgetary constraints.New measures to increase the effectiveness of the safeguards approach are being investigated that can be mutually beneficial to the facility operators and the IAEA.One of the key concepts being studied for application at future GCEPs is embracing joint use equipment for process monitoringof load cells at feed and withdrawal (F/W) stations.A mock F/W system was assembled at Oak Ridge National Laboratory (ORNL) to generate and collect F/W data from an analogous system.TheORNL system has been used to collect data representing several realistic normal process andoff-normal (including diversion) scenarios.Emphasis is placed on the novelty of the analysis of data from the sensors as well as the ability to build information out of raw data, which facilitates a more effective and efficient verification process.This paper will present the status of the current work to share data from operator equipment, introduce ideas on how to fully use the data to support information-driven inspections, and identify areas for future investigation.

Keywords: GCEP; IAEA; monitoring; process; integration

1. Introduction

Gas centrifuge enrichment plants (GCEPs) have been under International Atomic Energy Agency (IAEA) safeguards since the 1970s. The three primary safeguards objectives for enrichment facilities are (1) detecting diversion of declared material, (2) detecting excess production, and (3) detecting the production of higher than declared enrichments.Initially, the safeguards approach provided for inspectors to verify declared material balances but did not provide for inspector access to the centrifuge cascade halls to address the recognized potential of undeclared production. In 1980, the centrifuge technology holders convened the Hexapartite Safeguards Project (HSP) to develop the technical basis for which an IAEA safeguards approach could be established to detect undeclared production – specifically the production of highly enriched uranium (HEU).The HSP approach combined annual physical inventory verification (PIV) and routine interim inventory verification (IIV) inspections external to the cascade halls to verify material accountancy declarations withlimited-

frequency,unannounced access (LFUA) to the cascade halls to verify design and operations as declared. The initial LFUA inspection measures included visual inspection, nondestructive assay (NDA) on product header pipes, application of containment and surveillance, and the collection of samples for destructive analysis (DA).

As GCEP separative capacities grew beyond the original HSP design basis (1,000-tSWU/year) and as cascades increased in operational flexibility,new IAEA safeguards measures were incorporated into the safeguards approach as they became technically feasible and acceptable to the operators and inspectorates.In the early 1990s, the UK support program developed, tested, and later installed continuous header pipe enrichment monitoring systems with remote messaging to continuously monitor cascade product enrichment. Later in the 1990s, environmental sampling measures werealso added to the standard inspection approach.

In April 2005, the IAEA hosted a meeting of current enrichment technology holders (beyond those participating in the HSP)to evaluate the continued applicability of the HSP conclusions and to discuss the need and feasibility of incorporating additional safeguards measures as modern GCEPs increase in size and operational flexibility. During this meeting, the participants made a variety of recommendations. One of the recommendations was to evaluate the use of the "mailbox" approach on a case-by-case basis in connection with unannounced inspections and a short-notice,random inspection (SNRI) regime.[1] The mailbox declaration includes safeguards-relevant information such as the operational status of the feed and withdrawal (F/W) stations and, if in use, the identification (ID) of the cylinder inside. Periodically, the IAEA inspector arrives at the GCEP unannounced and accesses the mailbox and then verifies that the F/W stations are being used as declared. Subsequently, the IAEA evaluated this and other recommendations and eventually incorporated some of them into a new "Model" safeguards approach for GCEPs, which was released in 2006.

At the 2007 INNM Annual Meeting, a senior IAEA official presented the main elements of the new Model safeguards approach and stated that the Agency had decided to expand the use of randomized inspections (beyond those used for LFUA) to include SNRI for verification of declared feed operations and of nuclear material flows into, out of, and within the GCEP. In this presentation, the IAEA official confirmed that the safeguards objectives have remained unchanged from the HSP and outlined the main features of the model approach as follows:

1. an annual PIV;
2. design information verification;
3. nuclear material accountancy;
4. measures to confirm the absence of undeclared production; and
5. measures to detect enrichment higher than the declared maximum.[2]

2. Current efforts to investigate shared use of operator equipment

Given the three primary safeguards objectives and the five main features of the new approach,additional methods could further improve the effectiveness and efficiency of applied safeguards.

The U.S. Department of Energy's (DOE's) National Nuclear Security Administration (NNSA) is funding research on new measures that could be applied to GCEPs to improve the effectiveness of safeguards and increase the efficiency by decreasing the burden on the operator and the inspectorate.Several of the activities being sponsored by NNSA were described and demonstrated to NNSA,U.S. Government, and IAEA representativesat a January 2011 meeting hosted by the Savannah River National Laboratory (SRNL).Among the various laboratory participants, SRNL staff presented progress in developing an accountancy simulator consisting of a high-precision Wohwa Gyroscopic Accountancy Scale similar to the accountancy scales used at commercial GCEPs,Sandia National Laboratory (SNL) representatives presented approaches to authenticate operator data, and Oak Ridge National Laboratory (ORNL) personnel presented their findings related to the automatic analysis of process monitoring data from a F/W simulator.

3. Generating and collecting raw process data

The challenge of integrating data from disparate sources starts with an understanding of what data are available and then determininghow it all fits together.Currently, an IAEA inspector has access to the inventory listing, the inventory change report, and mailbox declarations.The inventory change report lists,among other things,the cylinder ID, the weight,and the enrichment level.Mailbox declarations are received periodically, e.g. daily,and identifythe specific cylinders in each respective F/W station.The work at ORNL funded by the U.S. DOE evaluateshow to correlateweight data taken from fully authenticated accountability scales and/or from the F/W stationswith existing information sources to enable the IAEA to achieve its objectives more efficiently.

The IAEA has expressed a desire for methods to detect undeclared production and to close the material balance period more frequently.To address these areas of interest, ORNL has focused on the automated analysis of load cell data from GCEPF/Wstations.ORNL has assembled a mock UF_6F/Wsystem in which water acts as a surrogate for $UF_6$to reproduce typical material handling operations.[3]This mock facilty has given ORNL the opportunity to generate and collect process load cell data and todevelop algorithms to analyze the data.The algorithms automatically identify process cycles and compute mass balances.This mock facility also demonstrated remote transmission of data to SRNL.

Part of the work being performed at SRNL involves analysis of tamper-indicating enclosures necessary for the shared use of the Wohwa Gyroscopic Scale used for accountancy measurements.If a larger plant contains many process scales (e.g., 50–100 or more), the cost to authenticate all of them could be prohibitive;however, it may be costeffective to improve authentication measures related to the high-precision accountability scales because there are only a few of these in any given facility, and all cylinders are weighed on one of these scales for the official declaration.

4. Raw data integration and evaluation

While the operator owns the accountability scale, the IAEA could independently collect data and fully trust it.However, this weight data linked to time does not directly reveal which cylinder is in a certain location at a specific time.Other information possibly available to the IAEA inspector would be necessary to build the complete picture.If the time the measurement was taken was included in the declaration, it could be used to find the corresponding weight in the IAEA dataset and compare the IAEA's weight value to the declared weight(see Figure 1 showing accountability scale data from the mock F/W system at ORNL shown with a mock declaration).This linkage would associatecylinder ID information to the accountability scale dataset.

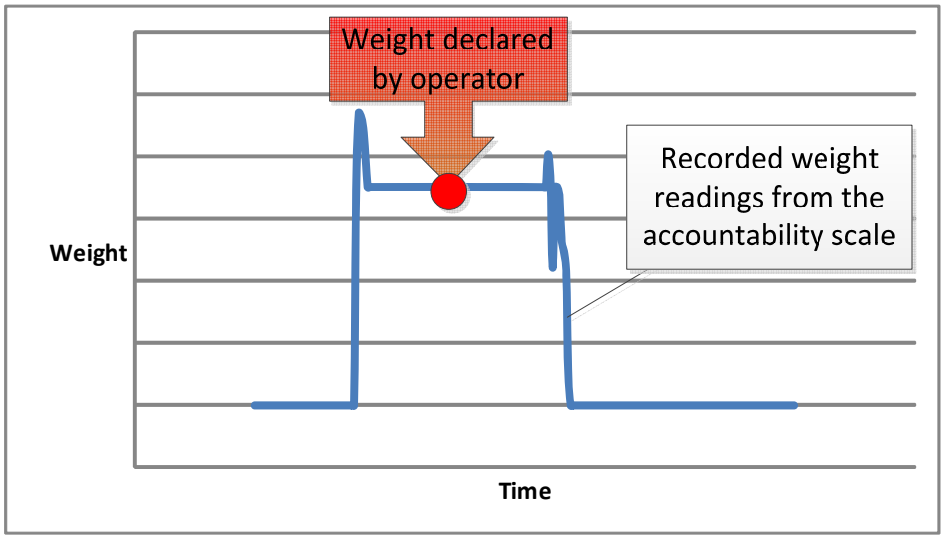

Figure 1: Example of fully authenticated accountability scale data.

Data from continuously monitoring process load cells could potentially be used to verify that only declared cylinders are processed and to close the material balance more frequently.Potentially, information from the authenticated accountability scale could be used to provide assurance in the recorded data from unauthenticated F/W stations.In a similar fashion as the accountability scale data, data from the F/W station load cells are collected without knowledge of the ID of the cylinder in the station.Mailbox declarations list which cylinder is in which F/W station for a specific time and could be used to linkF/W station process load scale data to a particular cylinder ID.As shown below in Figure 2, mailbox submissionsdeclare which cylinder is in a F/W station at a specific time, so it can be concluded that the cylinder with ID1 was in the station during the first filling, and the cylinder with ID2 was in the station for the second filling.

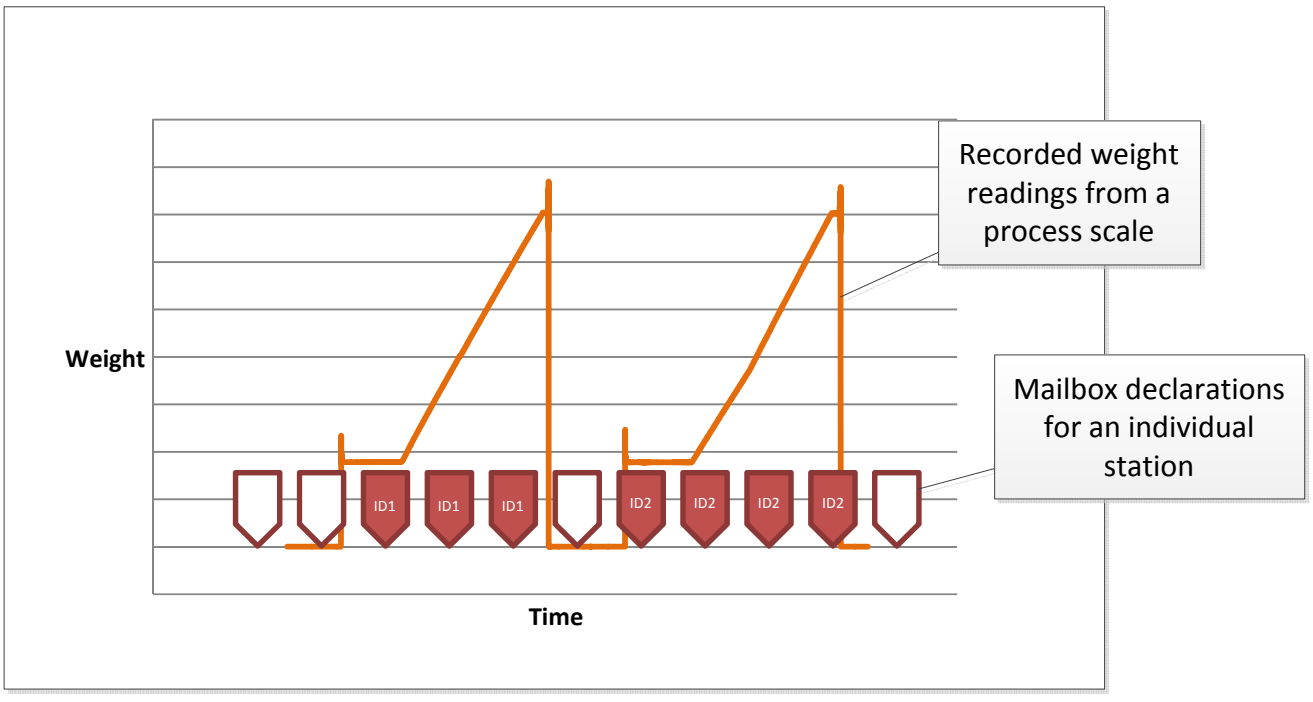

Figure 2:Integration of load cell data with mailbox declarations.

There is little confidence in the validity of a set of raw data presented independent of other information.However, confidence can be gainedwhen the “process” weights are compared to the weights from the fully authenticated accountability scale and with SNRI activities.[4]With the cylinder ID information extracted as demonstrated above, the authenticated accountability values for the full and empty weights can be compared and can add confidence in theweights extracted automatically fromprocess load cell data (see Figure 3). SNRI activities can add confidence that the F/W station was only used when declared in-use.

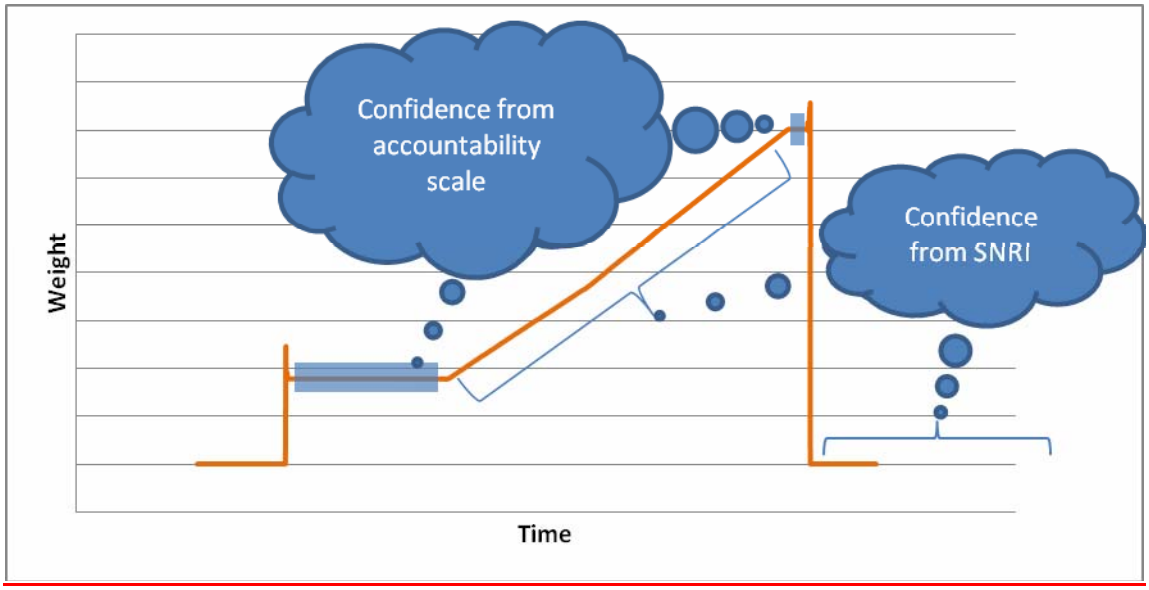

Figure 3: Confidence in full/empty weight gained from fully authenticated accountability scale.

Since the net mass fed intoor removed from the process is the parameter relevant to IAEA inspection efforts of material flow through the facility operations, and not the absolute weights, a bias in the process monitoring can be tolerated as long as the relative weight change remains consistent with the change declared.

5. Protecting sensitive operator information

Some operators have expressed concerns about the business and proliferation sensitivity of collecting and transmitting data from shared process equipment.While it has been proposed to delay transmission of process monitoring data to help protect an operator’s commercial and security interests, an alternative approach could rely on other process features (e.g.,a fully authenticated door switch or valve that must be open while UF_6 is flowing into or out of the process).A door switch could act as an on/off switch for recording F/W load cell data such that only when a F/W station door is open (or a valve is closed) would load cell data be released to and stored by the IAEA.Recall that the IAEA is concerned with verification of declared inventory and inventory changes, and detection of excess production. It is not necessarily concerned with the specific details of operator activities such as the exact shape of the plot showing the filling or emptying of a cylinder.If load cell values are collected only when a fully authenticated door switch indicates the station door is open, the inspectorate could still achieve its objectives of detecting diversion of declared material and detection of undeclared production.

Figure 4,obfuscates the data points that correspond to periods when the doors of aF/W station would be closed.However, the remaining data are sufficient to verify what material was deposited in the cylinder during this filling cycle. Correlating the empty and full weights to the values on the authenticated accountability scaleprovidesthe necessary information to calculate the weight of UF_6 delivered or removed from the process with some degree of confidence (see Figure 5).

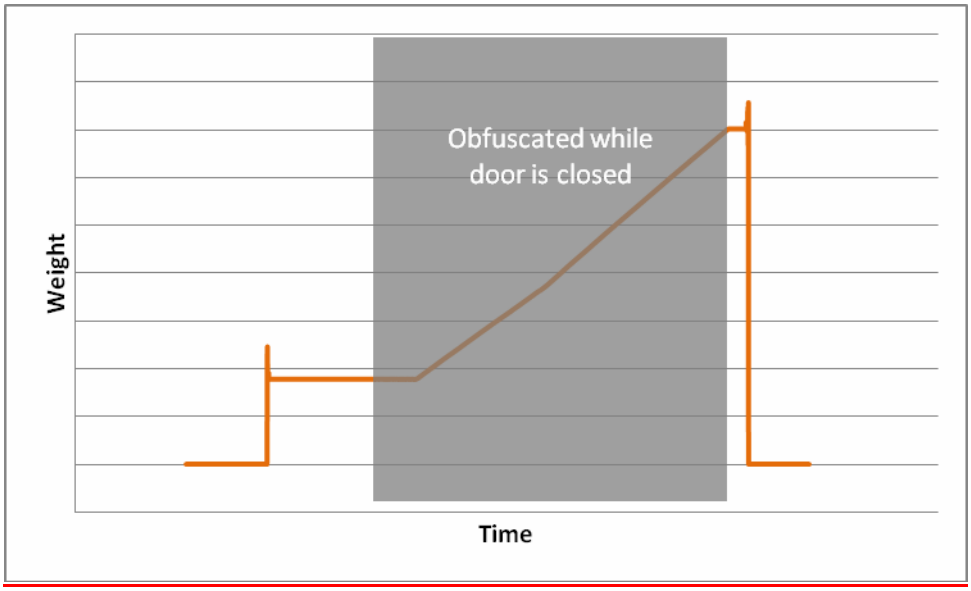

Figure 4: Load cell data obfuscated while withdrawal station door closed; confidence remains in the empty and full weights.

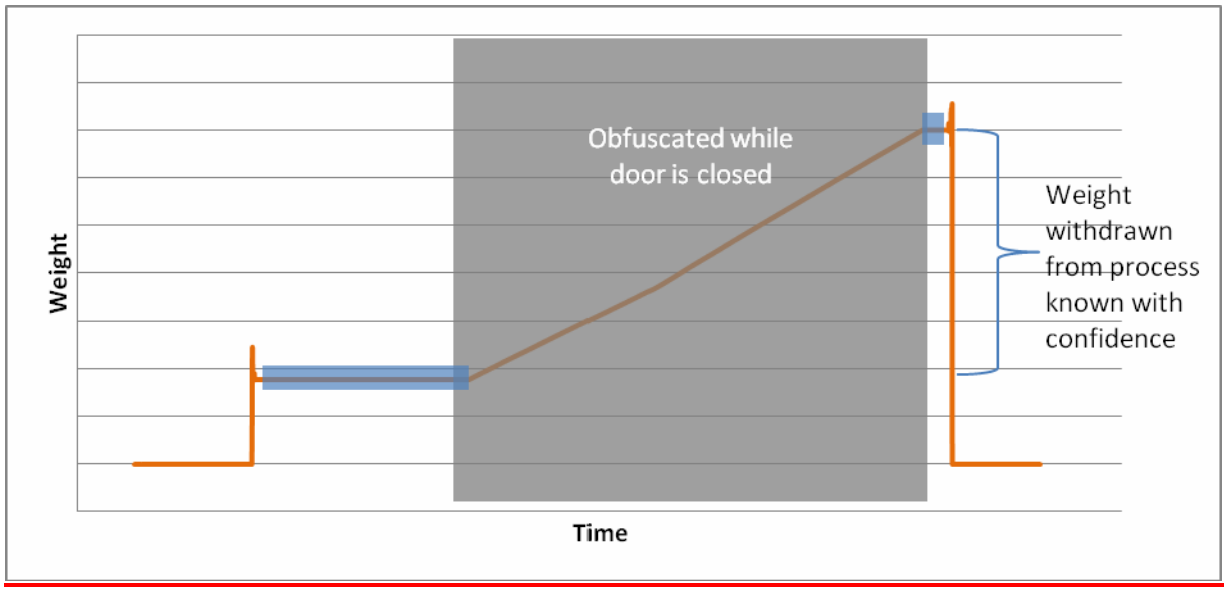

Figure 5: Obfuscated process monitoring data can still provide information for automated item counting and mass balance.

The concept of obfuscated data for in-process cylinders does have the drawback of potentially calculating larger daily fluctuations in the material unaccounted for (MUF).This is because the MUF calculationwould be based on the last value at the F/W stations before the data were obfuscated.Hence, the hardware switch discussed above may not fully meet the IAEA desirefordaily material balance calculations.In such cases,perhaps the operator may agree to allow data to be utilized to perform daily accountancy or other statistics calculations as long as the data from each station were not individually discernable and not permanently recorded.Another alternative may be to provide the current weight of cylinders in each F/W station along with the daily mailbox declarations to calculate the mass balance daily.In either of these arrangements, the mass balance could be computed and transmittedto the IAEA headquarters but would not provide the completedata series from individual stations.

6. Achieving safeguards objectives

The monitoring of process scales intends to significantly affect the second safeguards objective: detecting excess production.By integrating the data from several sources as shown above and automatically identifying inventory changes to and from the process, this new method can make great progress in providing assurance that only declared materials are introduced into the cascade through the declared F/W stations.Automated analysis of the available data should be able to culminate in computed inventory changes that could be compared to declared inventory changes.This could include cumulative information such as the number of cylinders fed, the number of cylinders withdrawn or the total weight of UF_6, U and ^{235}U fed or withdrawn. It could also include details-oriented review functionality to investigate specific cylinder information.

7. Using data to support information-driven inspections

The idea that all the relevant information at a GCEP should be collected and factored into the calculation ofwhen and how inspections are performed goes hand-in-hand with the IAEA's growing emphasis on objectives-based, fully information-driven safeguards.One way of applying this concept would be for all safeguards-relevant data to be combined into a confidence value, C; some example information streams are shown in Figure 6.This confidence value would reflect the IAEA's confidence that no undeclared activities or diversion of declared material was occurring and could be used to modify the random scheduling of a LFUA.Hypothetically, it could be updated weekly, or even daily, as new information becomes available and be used to modify the purely random probability of scheduling an unannounced inspection in a fully informationdriven approach.

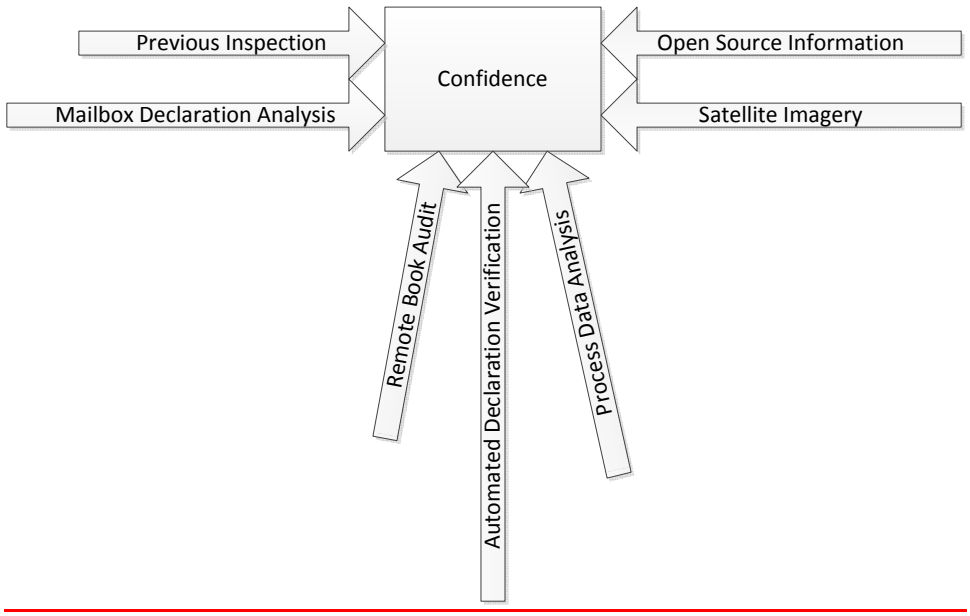

Figure 6: Confidence from ALL information sources.

8. Transmission of key performance indicators

Facility operators have been concerned that operational information should be available to the plant director before it is available to the IAEA. So while some information, such as state of health information for IAEA instrumentationcould be reported without delay, accountancy data may need to be delayed (e.g. for a few days, a week or even longer) to allow the facility staff to detect anomalies or accidents in operations before the IAEA would observe them. Perhaps some information may not be available at headquarters at all.Fully understanding what information needs to be reported and the frequency of transmission remains an area for future research.Additionally, significant effort would

need to be investigated to understand how to most appropriately incorporate different information sources to drive the inspection process.

9. Envisioned benefits to the application of international safeguards

Monitoring and automatically analyzing F/W station load data can help the IAEA more effectively detect excess production and undeclared cylinder operations.Beginning with automated comparison of declarations against process data, automated analysis can also provide for more frequent mass balance calculations.Currently, the mass balance is determined during the interim inventory verification (IIV)approximately monthly.However, as GCEP SWU capacity increases, the IAEA has expressed interest in an ability to compute the mass balance more frequently, potentially calculating it even on a daily basis.However, to obtain a daily material balance goal, new measures must be embraced to unify disparate remote data sources collected in nearreal-time.It must be stressed that the IAEA does not seek to get data just for the sake of getting more data but seeks to collect data in an effort to increase the efficiency and effectiveness of safeguards while, at the same time, benefitting the operator through potentially reduced IAEA personnel days of inspection, reduced residence time of product cylinders, reduced switchover activities, reduced number of destructive analysis samples, and a more timely resolution of anomalies before waiting for the next PIV.However the ability to perform more frequent mass balances is related to the frequency and timeliness of raw data and the ability to integrate disparate types of data.

10. Conclusion

Monitoring the F/W station load cells can help increase the effectiveness of detecting excess production, but only through integration of data from disparate sources can data from operator equipment begin to influence how inspections are carried out.While additional work is needed to understand how to best integrate data from disparate sources, F/W station load cell data can improve the effectiveness of detecting excess production today.

11. Acknowledgments

The authors would like to acknowledge the support provided by the Next Generation Safeguards Initiativethrough DOE/NNSA Office of Nuclear Safeguards and Security(NA-241) for this research.The authors would further like to acknowledge the comments from D. Hanks, B. Boyer, M. Curtis, K. Veal, and J. Damico.

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On-Line Enrichment Monitor for UF_6 GCEP

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Abstract:

This paper is a continuation of the Advanced Enrichment Monitoring Technology for UF_6 Gas Centrifuge Enrichment Plant (GCEP) work, presented in the 2010 IAEA Safeguards Symposium. Here we will present the system architecture for a planned side-by-side field trial test of passive (186-keV line spectroscopy and pressure-based correction for UF_6 gas density) and active (186-keV line spectroscopy and transmission measurement based correction for UF_6 gas density) enrichment monitoring systems in URENCO's enrichment plant in Capenhurst. Because the pressure and transmission measurements of UF_6 are complementary, additional information on the importance of the presence of light gases and the UF_6 gas temperature can be obtained by cross-correlation between simultaneous measurements of transmission, pressure and 186-keV intensity. We will discuss the calibration issues and performance in the context of accurate, on-line enrichment measurement. It is hoped that a simple and accurate on-line enrichment monitor can be built using the UF_6 gas pressure provided by the Operator, based on on-line mass spectrometer calibration, assuming a negligible (a small fraction of percent) contribution of wall deposits. Unaccounted-for wall deposits present at the initial calibration will lead to unwanted sensitivity to changes in theUF_6 gas pressure and thus to error in the enrichment results. Because the accumulated deposits in the cascade header pipe have been identified as an issue for Go/No Go measurements with the Cascade Header Enrichment Monitor (CHEM) and Continuous Enrichment Monitor (CEMO), it is important to explore their effect. Therefore we present the expected uncertainty on enrichment measurements obtained by propagating the errors introduced by deposits, gas density, etc. and will discuss the options for a deposit correction during initial calibration of an On-Line Enrichment Monitor **(**OLEM).

Keywords: UF_6, enrichment, OLEM, calibration, deposits

1. Introduction

A Continuous Enrichment Monitor (CEMO) was developed by AEA Technology, Harwell, UK in the early 1990's, with funding from the UK Safeguards Support Programme [1]. This was trialed at URENCO's gas centrifuge enrichment plants (GCEP's) at Almelo and Capenhurst [2] and has been in continuous operation on many of the enrichment cascades at

Capenhurst ever since. On each of these cascades, a CEMO is installed on a product pipe at the cascade outlet, before the first valve. The CEMO offers the following advantages:

- Rapid detection of Highly Enriched Uranium (HEU).
- Automated daily reporting of state-of-health to Euratom and IAEA HQ's.
- No possibility of removal of HEU before the instrument.
- Lightweight instrument, easy to fix to a UF_6 pipe.

However, the CEMO also has the following disadvantages:

- The use of a cadmium-109 source for the transmission measurement gives rise to high maintenance costs.
- The software is now rather old and consequently the monitor is not easy to use by safeguards inspectors.
- The UF_6 gas pressure at the location where CEMO is installed is classified information and so cannot be revealed to the safeguards inspectorates.
- The UF_6 gas pressure at the location where CEMO is installed is very low and thus the resulting measurement of ^{235}U enrichment is of low accuracy (but adequate for detection of enrichment above 20% on a plant licensed to 6%).

The Advanced Enrichment Monitor (AEM) being developed by Los Alamos National Laboratory (LANL) is similar to the Harwell-designed CEMO in that it uses a sodium iodide detector to measure gamma activity from UF_6 gas in a pipe and thus will continuously measure the ^{235}U enrichment. However, the AEM is designed to be installed at a different location in a GCEP, i.e. after the pumps, which are downstream of the cascade take-off valve consoles. The AEM has the following inherent advantages over the Harwell-designed CEMO:

- The UF_6 gas pressure at the location where AEM is to be installed is much higher that at the cascade outlet, and therefore the gamma radiation from the UF_6 is much stronger.
- The UF_6 gas pressure at the location where AEM is to be installed is not classified. This means that if a plant operator were prepared to disclose the pressure measurement to the safeguards inspectorates, then a cadmium-109 source would not be needed.
- On some GCEP's, the location where AEM would be installed is at a point where the take-off of several cascades has already been combined. This means with AEM, far less equipment would need to be installed in a plant than with CEMO.
- AEM should lead to a more accurate measurement of ^{235}U, at lower cost. This might even allow monitoring of tails flow, and not just product flow.

However, AEM does have the inherent disadvantage over CEMO in that it is designed to be installed downstream of the cascade take-off consoles, and so there would be opportunity to remove UF_6 before it reaches the instrument. The possibilities for an operator to defeat the instrument are thus higher than with CEMO. We must also recognise that CEMO is a proven instrument, with well over 15 years of routine use by Euratom and IAEA for safeguards verification. On the other hand, with the AEM, there is still much work to be done in testing the equipment in the field.

2. Basis of collaboration between LANL and URENCO

In May 2008, a meeting took place at URENCO's site at Capenhurst, UK, at which representatives of NNSA, LANL and ORNL proposed a trial at the URENCO enrichment plant of a Flow and Enrichment Monitor (FEMO), which was to be a combination of a ORNL-designed flow meter and a LANL-designed enrichment monitor. Over the ensuing period, it was decided to pursue testing of the LANL-designed enrichment monitor, which LANL

named the Advanced Enrichment Monitor (AEM). This is being carried out within the umbrella framework of a 2001 agreement between the US and UK governments for the development and implementation of nuclear verification technologies.

In January 2011, a "project arrangement" agreement was made between the US Department of Energy's National Nuclear Security Administration (NNSA), the UK Department of Energy and Climate Change, and URENCO for a trial of the AEM in URENCO's GCEP at Capenhurst.

Also in January 2011, LANL staff visited Capenhurst, to examine the location where AEM is to be installed in the plant and to carry out some initial measurements of a prototype of the AEM, which measured the ^{235}U signal only. IAEA technical staff took part as observers during this visit. The results obtained have not yet been analysed fully and are thus not given in this paper.

It is expected that the trial at Capenhurst will continue over the next one to two years, and that a summary of the results will be published to the wider safeguards community at appropriate international conference and workshops.

3. Potential uses of AEM

There are three **potential** uses by international safeguards inspectorates of an AEM in a GCEP:

- It could be used to rapidly detect the production of HEU. Compared with the current CEMO, it offers the potential advantages of reduced cost (both of initial investment and subsequent maintenance) and improved reliability.
- It might prove to be useful for verifying the ^{235}U enrichment of filled cylinders of enriched and depleted UF_6 produced by a GCEP. This capability might replace the current methods used by safeguards inspectors to verify enrichment of UF_6 in cylinders (viz. by portable gamma-spectrometers and by mass-spectrometry of samples of UF_6).
- There is the potential for installing a complete system on AEM's on all of the feed and take-off pipes in an enrichment plant and – when combined with load cell data on cylinder weighings – to calculate the mass balance continuously. However, the system required for this would be complex and might potentially reveal sensitive information on centrifuge cascade performance.

Note that URENCO has agreed only to the carrying out of a trial of the AEM, and in so doing has not committed to implementation of the AEM as a safeguards technique in any of its GCEP's.

There are two **potential** uses of an AEM by a GCEP plant operator:

- To provide a rapid warning if the enrichment of the product being made exceeds the enrichment limit for which the plant is licensed.
- To measure the enrichment of the feed to a GCEP, and the enriched and depleted material produced. This procedure could avoid the need for taking of samples (whether on-line or off-line) for analysis by mass-spectrometry. Use of on-line gamma-spectrometry would give a continuity of measurement not realised by mass spectrometry, but is unlikely to delivery the same standard of accuracy of measurement.

At this stage, it is far too early to say whether AEM will ever be suitable for use as a working instrument for any of the above applications. Above all, the equipment needs to be reliable, easy to install and operate and cost-effective. If these requirements can be met, then it is

likely that it could, in time, become an effective means of detecting HEU production in a GCEP.

4. Measurement conditions in unit header pipe

Figure 1 shows the changes in gas pressure, 235U count rate, gas attenuation and gas temperature that are anticipated to occur at the AEM location during the normal “fill and replace” cylinder cycle at a GCEP. These changes are used in some of the calibration options discussed below in section 5.2.

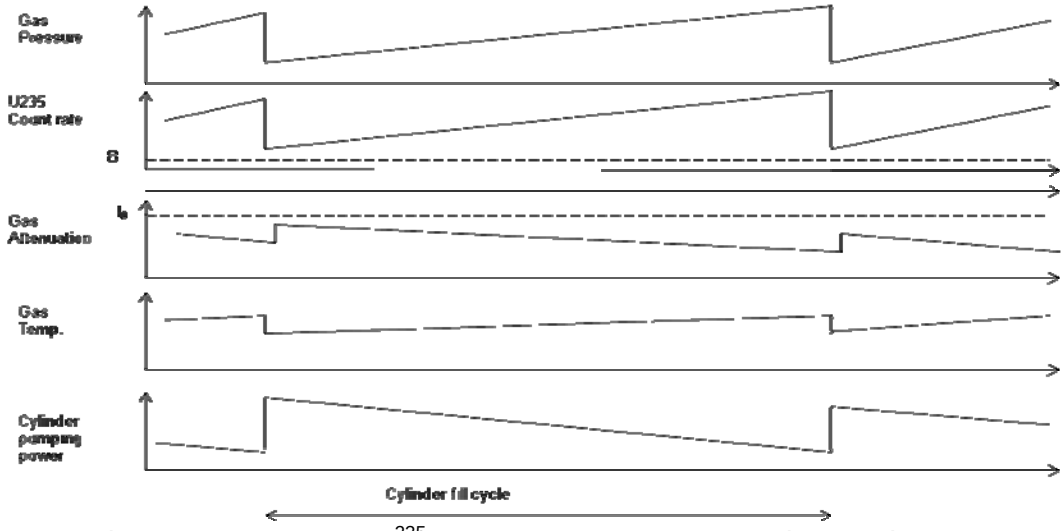

Fig. 1. Gas pressure, ^{235}U count rate, gas attenuation, and gas temperature as a function of cylinder filling.

5. Active enrichment monitor (186-keV spectroscopy + transmission based correction for UF6 gas density).

Correction for UF_6 gas density in this system is based on transmission measurements similar to those implemented in CEMO and blend-down monitoring system (BDMS) predecessor technologies. Unlike CEMO and BDMS, we have replaced the use of decaying ^{109}Cd or ^{57}Co isotopic sources with an X-ray generator and “notch filter” [3]. The prototype of the system being tested in the laboratory is shown in Fig. 2. The size and weight of the existing measurement head are large and may cause installation issues. A new design with reduced size weight and stress on the pipe is shown on Fig. 3.

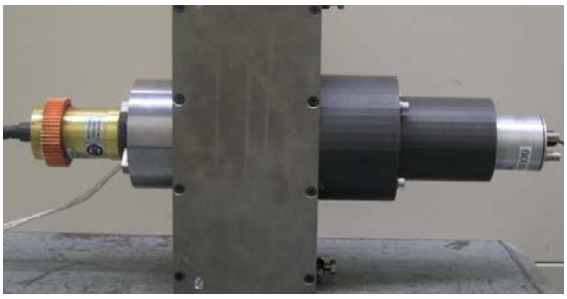

Fig.2. Existing active measurement head.

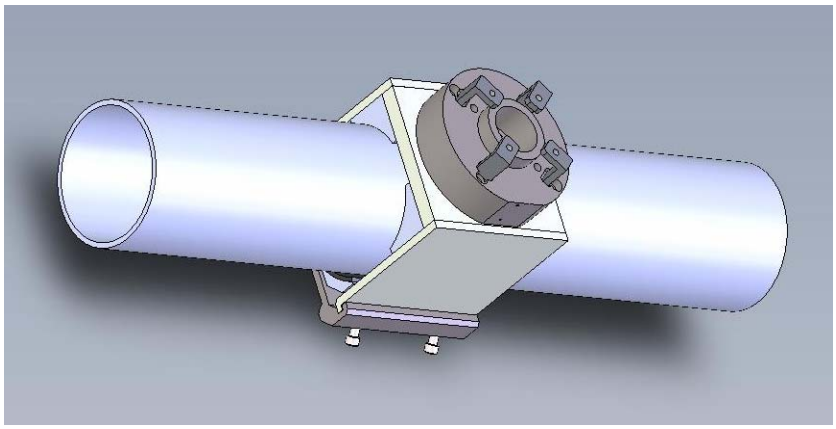

Fig. 3. Redesigned active measurement head.

5.1. Measurement method

The “active” method is based on two gamma spectroscopy measurements: a) intensity of 186-keV peak of ^{235}U and b) active transmission measurement for total UF_6 gas density.
The enrichment is calculated by the formula:

$$E(t) = K_{cal}^{a} \cdot \frac{R(t) - B}{ln\frac{I_0}{I(t)}} \qquad /1/$$

where
E = enrichment (%) of ^{235}U in the UF_6 gas,
K_{cal}^{a} = calibration constant,
$R(t)$ = count rate of ^{235}U, 186 keV, from UF_6 + background,
B = count rate of ^{235}U, 186 keV, from background,
$I(t)$ = count rate of the transmission source peak **with** UF_6 gas in the pipe, and
I_0 = count rate of the transmission source peak **without** UF_6 gas in the process pipe.

5.2. Calibration options

5.2.1. Classical (empty pipe) calibration

This calibration method has been used with CEMO and BDMS systems. It relies on three measurements:

- A gas off (empty pipe) measurement to determine the intensity of 186-keV peak due to wall deposits B and transmission peak intensity without gas attenuation to determine wall deposits contribution I_0.
- Gas on (calibration) measurement to determine transmission I_{cal} and 186-keV peaks R_{cal} intensities in the moment of calibration;
- Mass spectrometry measurements to determine UF_6 gas enrichment E_{cal} at the moment of calibration.

By solving Eq.1 for the calibration constant and substituting the obtained values from above measurements we can obtain the calibration constant K_{cal} .

$$K_{cal}^{a} = E_{cal}^{a} \cdot \frac{ln\frac{I_0}{I_{cal}}}{R_{cal} - B} \qquad /2/$$

Thus, Eqn. 1 is transformed into

$$E(t) = E_{cal}^{a} \cdot \frac{ln\frac{I_0}{I_{cal}}}{R_{cal} - B} \cdot \frac{R(t) - B}{ln\frac{I_0}{I(t)}} \qquad /3/$$

The classical calibration approach has proven operational history in CEMO and BDMS applications. We will use it as a reference during our field trial and testing new calibration approaches.

5.2.2. Calibration using gas pressure transients

This calibration approach is based on extrapolation of the intensities of the transmission and ^{235}U peaks at zero gas-pressure based on sets of measurements taken at different non-zero gas pressures, but with the same enrichment. This method is applicable when empty pipe measurement is not possible, or/and during the change of a UF_6 cylinder at withdraw station. We have previously experimented with this approach [9] for finding I_0 only. In this paper we describe an expanded calibration procedure for determining for both I_0 and B. Because the enrichment is the same for each pressure transient we can rewrite Eqn. 1 as follows:

$$E_1 = K_{cal}^{a} \cdot \frac{R_1 - B}{ln\frac{I_0}{I_1}} = E_2 = K_{cal}^{a} \cdot \frac{R_2 - B}{ln\frac{I_0}{I_2}} ;. E_3 = K_{cal}^{a} \cdot \frac{R_3 - B}{ln\frac{I_0}{I_3}} = E_4 = K_{cal}^{a} \cdot \frac{R_4 - B}{ln\frac{I_0}{I_4}} \text{, etc. /4/.....}$$

Where R_1, I_1,R_2, I_2 ; R_3, I_3,R_4, I_4...etc. R_n, I_n are the transmission and ^{235}U peak intensities during transients at different gas pressures. Eqn. 4 provides us following calibration options: a) Determine I_0 and B from three sets of measurements or b) Determine I_0 and B based on fit to all available data points.

This approach could be applicable for measurement on unit header pipe where pressure transients can be used for generation of calibration data.

6. Passive enrichment monitor (186-keV spectroscopy + pressure based correction for UF_6 gas density)

This system relies on pressure-based correction for UF_6 gas density, where the facility operator provides the pressure information, and therefore can use simpler hardware. The system concept and design architecture is described elsewhere [4, 5]. The installed dual-detector head is shown in Fig 4.

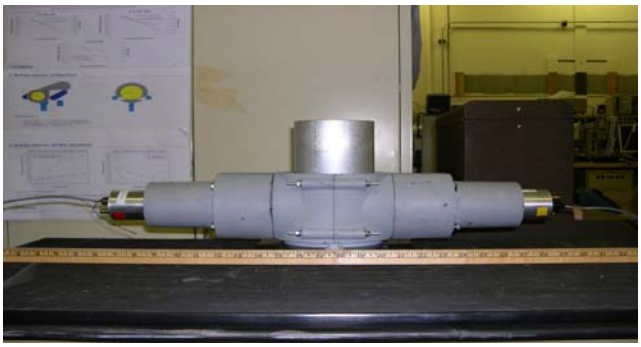

Fig. 4. Two passive detectors in the “face-to-face configuration deployed at Capenhurst.

6.1. Measurement method

For a “passive” measurement, the measurement for UF_6 gas density is provided by gas pressure and temperature according to the gas law:

$$\rho = \frac{p}{R_{UF6}T} \qquad /5/$$

Where:
p is the gas pressure
ρ is the gas density
R_{UF6} is the UF_6 specific gas constant

Because the differences from the ideal gas law are small for UF_6 [6], we use an ideal gas dependency $p = \rho RT$ in our analysis and will address any nonlinearity in our calibration.
The enrichment is calculated as:

$$E(t) = K_{cal}^{p} \cdot \left(R(t) - B\right) \cdot \frac{T(t)}{p(t)} \qquad /6/$$

Where:
E = enrichment (%) of ^{235}U in the UF_6 gas,
K_{cal}^{p} = calibration constant for passive measurement (that includes the universal gas constant R,
$R(t)$ = count rate of ^{235}U, 186 keV, from UF_6 + background,
B = count rate of ^{235}U, 186 keV, from background,
$p(t)$ = UF_6 gas pressure in the pipe
$T(t)$ = UF_6 gas temperature

The operator’s pressure gauges provide accurate and prompt information for UF_6 gas pressure, but the gas temperature is unknown.

6.2. Dealing deal with unknown gas temperature

Because the pressure in the header pipe rises during the cylinder fill cycle, the gas temperature also will change, thus it has to be considered as an unknown variable, rather than unknown constant like I_0 and B. The correction for unknown gas temperature is the main

challenge for implementation of an accurate passive enrichment monitoring system. Because the gas temperature is correlated with gas pressure, direct empty pipe or gas pressure transients calibration methods similar to an active system is not feasible for a passive system. Because both passive and active methods share the same measurement for concentration of ^{235}U, the active transmission measurement can be used to calibrate a passive, pressure-based measurement for UF_6 gas density. This would involve side-by-side measurement using both active and passive systems combined with a proven thermodynamic model of gas behaviour within the unit header pipe. Alternatively, it may be possible to determine the relationship between gas temperature and gas pressure using the passive system during a period when it is known by URENCO that the enrichment level of the UF6 is stable.

7. Conclusions

- An Advanced Enrichment Monitor is being developed by LANL and tested on a URENCO GCEP.
- The AEM has the potential to offer advantages in cost, reliability and accuracy compared with the existing CEMO.
- Both passive and active enrichment measurement methods are used in AEM development.
- Pressure transients could be used for self-calibration.
- The measurement environment at the unit product header pipe has been described.
- Unknown UF_6 gas temperature variations present a major challenge for implementation of the passive enrichment method.
- The temperature variation is related to the enrichment process, and therefore can be addressed by joint R&D efforts.

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23 Knowledge Management and Training

Technical convergence of nuclear safeguards, nuclear forensics and nuclear security

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Abstract:

From the first implementation of safeguards agreements, measurements of nuclear material have formed the backbone of physical verification. The introduction of strengthened safeguards and the ratification of additional protocols (INFCIRC 540) led to the need for more investigative analytical measurements, similar to those implemented for the analysis of nuclear material intercepted from illicit trafficking. The requirement of nuclear forensics, to disclose additional information inherent to nuclear material, triggered the transfer of analytical techniques from the environmental area, from materials science and from the geological and cosmological areas to the safeguards community. This transfer of expertise significantly contributed to the identification of the sources of nuclear materials and the industrial processes to which they have been exposed.

The physical security of nuclear material is the responsibility of the State that owns the material. However, with increasing concern over non-State actors, it is important for the wider community to work in cooperation with States to which intercepted nuclear materials might be attributed. During the ESARDA annual meeting 2010, the ESARDA Working Group Techniques and Standards for Destructive Analysis (WG DA) adopted a new Objective in its Action Plan 2010-2012: to emphasise the technical convergence of nuclear safeguards, nuclear forensics and nuclear security by looking at available and new methodologies that serve all three purposes. The WG DA organised dedicated workshops on advancements and applicability of analytical techniques to read signatures in nuclear material and environmental samples, with participation beyond the safeguards community. The aim was to bring together experts from safeguards, nuclear forensics, earth sciences, and particularly from industry. Most recently, immediately prior to this symposium, the ESARDA WG DA held a dedicated workshop on direct analysis of solid samples using Laser Ablation-Inductively Coupled Plasma Mass Spectrometry (LA-ICP-MS), hosted by the Hungarian Atomic Energy Authority (HAEA). Furthermore the WG DA interacts with the WG NA/NT (Novel Technologies / Novel Approaches) to evaluate synergies.

The WG DA contributed to the revision of the International Target Values for Measurement Uncertainties in Safeguarding Nuclear Materials (ITVs) and a Workshop on Uncertainties in Nuclear Measurements is scheduled for the end of this year. These contributions provide operators and inspection authorities with important information on the capabilities and expected performance of analytical techniques. The WG DA has developed DA and forensics modules for training and education that are regularly presented at the ESARDA academic course on Nuclear Safeguards and Non-Proliferation and the Belgian Nuclear Higher Education Network (BNEN) advanced course on safeguards, which are recognised by the European Nuclear Higher Education Network. Additional courses in France and Sweden are scheduled for this year. Dedicated training courses in the area of nuclear forensics are offered at the Institute for Transuranium Elements (ITU). Moreover, a "European Nuclear Security Training Centre" is being established at the ITU, with the support of DG HOME. The European nuclear security training centre (EUSECTRA) will offer a broad spectrum of training courses including detection of and response to nuclear security incidents and nuclear forensics. Within the WG DA, ESARDA is currently keeping abreast of methodologies applicable in the fields of nuclear safeguards, security and forensics.

Keywords: nuclear safeguards, nuclear forensics, nuclear security, destructive analysis, education and training

1. Introduction

In the COUNCIL DECISION 2010/212/CFSP of 29 March 2010 relating to the position of the European Union for the 2010 Review Conference of the Parties to the Treaty on the Non-Proliferation of Nuclear Weapons, the European Union confirmed its view to strengthen the NPT as the cornerstone of the international nuclear non-proliferation regime. The three pillars of the NPT are non-proliferation, disarmament and peaceful uses of nuclear energy. Nuclear security constitutes a major challenge for the 21st century, as recognised at the 2010 nuclear security summit in Washington DC, USA. The three pillars of nuclear security are prevention, detection and response. Nuclear security in the broad sense includes countering nuclear illicit trafficking, proliferation resistance and physical protection. Bringing this concept one step further nuclear safety and security can be seen as two sides of the same coin. The triple concept SSS (Safeguards, Security, and Safety) is converging more and more towards the idea of internationally binding security and safety standards. The COUNCIL DIRECTIVE 2009/71/EURATOM of 25 June 2009, establishing a Community framework for the nuclear safety of nuclear installations states that *information to the regulation of nuclear safety is made available to the general public, provided that this does not jeopardise other interests such as, inter alia, security, recognised in national legislation or international obligations*. After the natural disaster that hit Japan in March 2011, the EU stated in the EUROPEAN COUNCIL conclusion from 24/25 MARCH 2011 that the highest standards for nuclear safety should be implemented and continuously improved. This applies also to nuclear security, as emphasised by the President of the European Council, Herman van Rompuy, on behalf of the EU at the Nuclear Security Summit *'the EU, with its large multinational nuclear industry, has a particular interest in ensuring that the peaceful uses of nuclear energy take place with the highest standards of nuclear safety, security and non proliferation'* [1]. There are differences between nuclear safeguards and security considering their political and legal aspects. Nuclear safeguards is based on international agreements such as INFCIRC 193 and Euratom regulation 302/2005, and the subsequent introduction of strengthened safeguards and the ratifying of additional protocols (INFCIRC 540). Nuclear security is the responsibility of regulatory bodies at the national level but with increasing concern over non-State actors inter-States cooperation is important.

Seen from the technical point of view nuclear safeguards includes fissile material control whilst nuclear security includes physical security of nuclear material, identification of possible signatures, monitoring, sample analysis and identification of material and design. The latter is also part of nuclear forensics which deals with consistency of information, coherence between materials or samples, conformity of findings with declared processes and comparison of external and internal data. Since the introduction of strengthened safeguards more investigative analytical measurements, similar to those implemented for the analysis of nuclear material intercepted from illicit trafficking, are also applied to safeguards. To bring the technical convergence forward exchange of expertise between the different communities is indispensable together with advancements in analytical and technical techniques.

2. Technical convergence of nuclear safeguards, forensics and security

The ESARDA Working Group Techniques and Standards for Destructive Analysis (WG DA) adopted a new objective in its Action Plan 2010-2012 looking at available and new methodologies that serve nuclear safeguards, nuclear forensics and nuclear security. The ESARDA WG DA strategy to address this challenging objective of technical convergence is to establish exchange beyond the safeguards community on dedicated technical topics relevant to all three fields, to participate in respective consultant or expert group meetings and to support ESARDA's educational role in reaching the general public. This strategy is perfectly in line with the recommendations of the ESARDA Reflection Group 2010 that ESARDA should expand its scope towards the convergence of nuclear safeguards, nuclear security and nuclear forensics/illicit trafficking.
One main focus of the WG DA is supporting activities for the development and improvement of methods for the determination of nuclear signatures in environmental and "special" samples. Isotopic "fingerprinting" is needed for attribution of intercepted materials and for verification of the correctness and completeness of a State's declarations, particularly in view of the detection of any undeclared material or activities. Recently, under the initiative and coordination of IRMM, four key nuclear mass spectrometry laboratories – namely the Institute for Reference Materials and Measurements (IRMM), the Institute for Transuranium Elements (ITU), the International Atomic Energy Agency-Safeguards Analytical Services (IAEA SGAS), and the US Department of Energy-New Brunswick Laboratory

(DOE-NBL) - published an article on the development of the Modified Total Evaporation technique (MTE) applied for sample analysis in nuclear safeguards, nuclear forensics and other disciplines like geo-and cosmo-chemistry [2]. MTE is a new tool for thermal ionization mass spectrometry in nuclear safeguards and geochemistry developed as a method for accurate measurements in particular for minor isotope ratios of uranium in nuclear material, and in combination with a multi-dynamic measurement procedure, for plutonium in environmental samples. The MTE method provides a measurement performance which is superior to the present IAEA requirements, enabling more detailed conclusions from sample data for source attribution of samples. In a harmonised approach the MTE method was successfully integrated for routine use at all contributing laboratories and has subsequently been applied on measurements of samples in safeguards and forensics. In addition the application of MTE goes beyond safeguards since it provides a significant advantage for low mass elements like boron or calcium allowing an unprecedented level of accuracy for isotope ratio measurements. This is an outstanding example of joint advancements in measurement sciences of international reference and safeguards laboratories that exchange expertise via the ESARDA WG DA platform.
Advancements in low-level analytical techniques serve nuclear safeguards, nuclear forensics and nuclear security purposes. An example is the successful determination of plutonium isotope ratios in environmental samples from different geographic origins and from Chernobyl, through application of applying state of the art Thermal Ionization Mass Spectrometry (TIMS) in combination with multiple ion counting (MIC) and filament carburization [3]. Due to this developed procedure, data were acquired with lower uncertainties not only for the $^{240}Pu/^{239}Pu$ major isotope ratio but also for the $^{241}Pu/^{239}Pu$ and $^{242}Pu/^{239}Pu$ minor isotope ratios, which is particularly important for fingerprinting of environmental samples. This information reveals different sources of plutonium contamination in the environment and can help to distinguish if the observed contamination in collected samples originates from global fallout or from an accident like in Chernobyl. This technique could also be applied to environmental samples collected around the Fukushima site.

2.1. Dedicated ESARDA WG DA workshops

Research towards advancements in techniques enabling the determination of nuclear signatures is not exclusively of interest to the safeguards and security sector but also to the geochemistry, environmental and earth sciences community. Therefore the WG DA organises dedicated workshops on advancement and applicability of analytical techniques to read signatures in nuclear material and environmental samples. The aim is to bring together experts from safeguards, nuclear forensics, earth sciences, geochemistry and particularly from industry. This stimulates and encourages the exchange of information beyond the safeguards community using the ESARDA WG DA as a platform. Successful examples of WG DA workshops from the past are the *Workshop on Measurements of Minor Isotopes in Uranium* and the *Workshop on Measurements of Impurities in Uranium*. The outcome of the workshops is published in the ESARDA bulletin to communicate technical issues on safeguards, forensics and security to a broader public [4, 5].

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2.1.1. Revision of the International Target Values 2010

The ESARDA WG DA is promoting use of the International Target Values for Measurement Uncertainties in Safeguarding Nuclear Materials (ITVs), which are uncertainties to be considered in judging the reliability of analytical techniques applied to industrial nuclear and fissile material subject to safeguards verification. The concept of ITVs was originally conceived in 1979, by the WG DA, and has matured gradually over the years, eventually taken over by the IAEA in the early 1990's. They have been increasingly utilised in international Safeguards, not only as a measure of performance for facility operators and measurement laboratories, but also as a reference for Safeguards Authorities to use as a reasonable (minimum) requirement. The ITVs are a reference of the quality of state-of-practice measurements achievable in nuclear material accountancy. They have to be fit for purpose and achievable under routine working conditions.
In 2010 the ESARDA WG DA together with the NDA working group organized dedicated meetings to revisit the ITVs providing significant input to the ITV2010 review done by the ESARDA WG DA and NDA, IAEA, ANSI/INMM 5.1, Japanese ITV-2010 Expert Group, ABACC, EURATOM, ISO TC 85/SC5 and E. Kuhn as consultant. A major achievement was to include upon the recommendation of the ESARDA WG DA relative combined standard uncertainties together with a new chapter on *GUM and*

the Use of ITVs by Measurement Laboratories. This new chapter in the ITV2010 document establishes the link between the current approach of laboratories evaluating their measurement uncertainties according to GUM and the safeguards evaluators needs to identify when an operator-inspector difference exceeds a certain limit. The ITV2010 have been published in November 2010 and are also accessible via the ESARDA WG DA web-site [6].
The concept of ITVs, in setting fit-for-purpose achievable performance criteria for measurement laboratories, has the potential to be also applicable to environmental bulk and particle sample analysis. Similar concepts have already been applied in other areas, such as water, food and clinical analysis where important decisions are based on measurement results and compliance with legal or regulatory limits must be demonstrated.

2.1.2. Workshop on direct analysis of solid samples using LA-ICP-MS

One of the most powerful tools to detect undeclared nuclear activities is the particle and bulk analysis of environmental samples. Bulk analysis of the collected swipe samples using Thermal Ionisation Mass Spectrometry (TIMS) or Multi Collector-Inductively Coupled Plasma-Mass Spectrometry (MC-ICP-MS) may be used, whilst methods for investigation of single particles can improve significantly the detectable amount of nuclear materials. Nowadays, alongside other techniques (e.g. Secondary Ion Mass Spectrometry, SIMS), ICP-MS combined with laser ablation (LA) sample introduction allows the direct investigation of the isotopic composition of uranium and transuranium elements in single particles. The ESARDA WG DA in close collaboration with the Hungarian Atomic Energy Authority (HAEA) is organising a dedicated workshop on *Direct Analysis of Solid Samples Using Laser Ablation-Inductively Coupled Plasma-Mass Spectrometry (LA-ICP-MS)*, held in conjunction with the ESARDA Symposium in May 2011. The workshop aims to explore the potential of LA-ICP-MS for safeguards, non-proliferation, nuclear forensics and other applications. The ESARDA WG DA and the HAEA succeeded in gathering distinguished speakers and participants for this workshop from safeguards, chemistry, geology, earth sciences and industry to exchange views and experience in this area. The final report of this workshop is expected to be published in the ESARDA Bulletin, Dec. 2011.

2.1.3. Workshop on Uncertainties in Nuclear Measurements

One of the objectives of the ESARDA WG DA is to address measurement problems arising from new challenges in safeguards and in related areas. Intrinsic to all measurement results is that they are the basis of any conclusion to be drawn from an analytical process therefore they need to be reliable, traceable and accurate. Particularly when investigating the applicability of new techniques and instrumental methods to address safeguards and security issues inter-comparability of results between methods and laboratories are of major importance. Deriving a quantitative statement of the measurement uncertainty associated with a measurement result is thus crucial. The WG DA will organise in cooperation with the IAEA and in conjunction with its annual working group meeting a dedicated Workshop on *Uncertainties in Nuclear Measurements* at the IAEA in Vienna from 8-10 November 2011. Topics amongst others will be uncertainty estimation from state-of-the-art to state-of-the-practice measurements with emphasise on transparency in calculation and estimation. This workshop provides a platform for exchange between reference measurement institutes, safeguard laboratories, nuclear and environmental material analysts and in particular operators. The aim is to investigate the major contributions to the final measurement uncertainties depending on the material and technique applied, and to compare and discuss the different approaches in uncertainty estimation. The ITV2010 as well as the consistency of measurements carried out by nuclear laboratories and by operators with the GUM approach will also be an important topic. The final report of this WS is expected to be published in the ESARDA Bulletin, June. 2012.

3. Training and Education

Chapter I of the EURATOM treaty concerning the promotion of research and the European Council conclusion on *the need for skills in the nuclear field* set out the legal framework for putting efforts in maintaining and transferring the expertise in the nuclear field from one generation of engineers and researchers to the next [7]. The WG DA strongly supports ESARDA in fulfilling an educational role in the nuclear field and also in reaching the general public. Members of the WG DA have for some years

been active in the area of education and training in safeguards & non-proliferation. Technical sheets on analytical techniques, reference materials and quality control tools are accessible to the public via the ESARDA library web-site [8].The ESARDA WG DA cooperates closely with the WG on Training and Knowledge Management and has developed modules on destructive analysis and forensics. Recently other communities with a background in nuclear safety in environmental sciences in political sciences or economy, and even in social sciences like peace research, showed more and more interest in learning about the relevant treaties in nuclear non-proliferation, the verification strategies, the applied analytical techniques and nuclear security issues linked to nuclear forensics. Especially during this year's courses, a number of questions triggered by the Fukushima accident concerning reactor safety, radiation doses and nuclear signatures in the environment were asked by the participants. These topics are beyond regular safeguards courses, although some answers can be given via examples on the application of techniques to assess significant parameters that are similar in safeguards, security and safety. Furthermore the role of the IAEA in safeguards, security and safety was of major interest to the training course participants. It seems, that in the field of training, the technical convergence is starting to be successfully implemented as can be seen from the following examples.

3.1. ENEN training courses on safeguards

The destructive analysis and forensics modules are regularly presented at the ESARDA academic course on Nuclear Safeguards and Non-Proliferation for students and young professionals on Nuclear Safeguards and Non-Proliferation and from part of the course syllabus [9]. In 2009 the essay on *Fingerprinting of Nuclear Material for Nuclear Forensics* and, in 2010, the essay on *Destructive Analysis: Effective Analytical Support to Nuclear Safeguards and Non-Proliferation*, were selected as Best student's paper of the respective ESARDA courses and published in the ESARDA Bulletin [10, 11]. The 8th ESARDA COURSE will be the first one on the initiative of Sweden, to be held outside the Joint Research Centre (JRC), in Uppsala, from September 12th – 16th, 2011. Furthermore an additional ESARDA course is planned to be held in France. The WG DA modules will be presented at both courses. The module on destructive analysis has twice been presented at the BNEN advanced course on safeguards in Belgium, also recognised by the European Nuclear Higher Education Network [12, 13].

3.2. Use of Reference Materials and the Estimation of Measurement Uncertainty

IRMM regularly provides a training course on the *Use of Reference Materials and the Estimation of Measurement Uncertainty*. Since this course provides participants with the theoretical basis for the estimation of measurement uncertainty, establishment of traceability, and the proper selection and use of reference materials it is applicable to all measurement fields. The aim is to use reference materials to achieve true traceability of measurements, proving accuracy of methods and demonstrating proficiency of laboratories. The course strongly emphasises practical application of the theoretical concepts presented during the lectures. Dedicated exercises are undertaken by the participants in small groups with support from a trainer. Although the course layout was originally set for laboratory managers, practitioners in analytical laboratories and ISO 17025 auditors from the non-nuclear field, it is now also very much appreciated by nuclear inspectors from Euratom Safeguards coming from the field of non-destructive analysis. The next course will be held from 12-13 October 2011 at IRMM [14].

3.3. Nuclear forensics training

Dedicated training courses in the nuclear forensics area are offered at the Institute for Transuranium Elements. Training toics range from conceptual approaches in nuclear forensics, to modelling and calculations, to practical hands-on training in the laboratory.

The nuclear science web portal NUCLEONICA (http://www.nucleonica.net/index.aspx) contains a number of modules and applications that are relevant to both nuclear safeguards and nuclear security applications. This includes a gamma spectrum generator, shielding calculations, decay calculations or simple reactor code calculations (webKORIGEN). Specific Training Courses on Illicit Trafficking and Radiological Consequences with Nucleonica address this area (typically once per year). The conceptual approaches in nuclear forensics are discussed in seminars on "nuclear forensics

awareness". The focus of these seminars is on the interaction between different agencies involved in handling cases of illicit trafficking. A clear definition of roles and responsibilities is required and particularly those authorities who are less familiar with nuclear forensics (e.g. police, radiation protection, environmental protection) acquire basic knowledge of the protocols to be followed in order to preserve nuclear forensic evidence. Much more technical and laboratory oriented training courses are offered for technical experts from national laboratories. These are typically provided to small groups (1-3 persons) and tailored to specific technical topics. Examples of technical disciplines in which training is offered are: electron microscopy; mass spectrometry; radiochemical separations; alpha spectrometry; and age dating of nuclear materials. This training is carried out upon request.

In the same context, ITU also carries out "joint analysis exercises" where nuclear material from an EU Member State or other States (e.g. from a seizure or test materials) are analysed in the laboratories of ITU with participation of national experts. This offers on the one hand the opportunity to provide hands-on training to the participating national experts. On the other hand, it allows the exercising of all the procedures - from transport of the sample to drafting of the report - that would also be required in a real case. Recently, a "joint analysis" exercise was carried out with Hungary and a more detailed report is provided at this conference [15].

3.4. EUSECTRA

Training of responders in nuclear security has been identified as a high priority on the European level. The JRC is creating, on the request of the Directorate General for Home Affairs, a *European nuclear security training centre* (EUSECTRA), at ITU, to complement national training efforts. The courses including detection of and response to nuclear security incidents will be split in a theoretical part and practical sessions. It is envisaged to provide training under realistic scenarios. To this end, a detection area using both scintillator detectors and spectrometric detection systems is set up along a road. This will mimic situations that may occur at border crossing stations or other nodal points. The objective is to provide advanced training, allowing the trainees to gather hands-on experience in the detection and categorization of nuclear material. Moreover, a dedicated laboratory space is being set up where the trainees will be able to use different types of hand-held or portable equipment to measure radioactive or nuclear material in order to experience the capabilities and limitations of the instrumentation. EUSECTRA will also include training on nuclear forensic awareness, on establishing core capabilities in nuclear forensics and on the development and implementation of a national response plan.

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4. Conclusion

The ESARDA WG DA sees in contrast to the relevant differences of political and legal nature between nuclear safeguards, forensics and security, especially in the advancement and applicability of analytical techniques. It has been shown that operating the ESARDA WG DA as an interface and exchange platform is an effective strategy towards technical convergence of nuclear safeguards, forensics and security. The organisation of dedicated workshops and active participation in training courses on topics relevant to these three fields create a public awareness of the synergies beyond the safeguards community and opens possibilities for cooperation in research and development. One could summarise that nuclear safeguards is about the complete, correct and comprehensive picture of a State's nuclear activities; nuclear forensics about the identification of the origin and intended use disclosing additional information inherent to nuclear material; and nuclear security about prevention/detection/response of theft, sabotage, unauthorized access, illegal transfer of nuclear/radioactive materials and associated facilities. Verification, detection, conformity and response are based on reliable measurement results with appropriate quality control tools as prerequisite and on advanced measurement techniques and instrumentation. This is exactly the point where the similarities between nuclear safeguards, nuclear forensics and nuclear security start to balance out the differences they have at the political level. Table 1 attempts to give an overview on how nuclear safeguards, nuclear forensics and nuclear security are linked on the technical level, including the necessity of information from other sources. Main emphasis is put on training and education which is beneficial for researchers, engineers, scientists, and (young) professionals from all three fields. Training courses and publicly available education materials enable and stimulate cooperation and, as a spin-off, enhance the competitiveness of researchers in the job market. Therefore, to promote the technical convergence of nuclear safeguards, nuclear forensics and security is not only in line with the EU flagship 2020 of sustainable growth, but supports particularly the EU flagship initiative *Youth on the move* [16]. Within the WG DA, ESARDA is currently keeping abreast of methodologies applicable in the fields of nuclear safeguards, security and forensics.

Table 1: Technical convergence of nuclear safeguards, nuclear forensics and nuclear security

Technical convergence of nuclear safeguards, nuclear forensics and nuclear security					
Scientific Disciplines : Chemistry, Physics, Material science					
Analytical and technical tools					
	Nuclear material analysis Verification of non-diversion	Environmental sample analysis Detection of undeclared activities	Seized/collected material analysis Consistency, Coherence, Conformity of information, materials and processes	Metrological quality control tools Method development, method validation, QC/QA	Other sources of information
	Amount content, isotopics	Bulk and particle analysis; isotopic fingerprint	Bulk and particle analysis; isotopic fingerprint; anionic and metallic impurities, microstructure, "age" – last separation date	Certified Reference Materials (CRMs) Interlaboratory Comparisons (ILCs)	Close cooperation with data analysts, police, governments,…
Nuclear Safeguards Complete, correct and comprehensive picture of a State's nuclear activities	✓	✓		✓	
Nuclear Forensics Identifying origin and intended use using information inherent to the (nuclear) material		✓	✓	✓	✓
Nuclear Security Prevention/Detection/Response of theft, sabotage, unauthorized access, illegal transfer of nuclear/radioactive materials and associated facilities			✓	✓	✓

5. Acknowledgements

The authors would like to acknowledge the efforts of all the ESARDA WG DA members in the organisation of the dedicated WG DA workshops and their input to the training events. The scientific input and expertise of all the working group members, and also of invited experts, are highly appreciated. Furthermore the authors would like to thank the members of the ESARDA Reflection Group 2010 for fruitful discussions with concerning nuclear safeguards, forensics and security matters.

6 Legal matters

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Systematic classification of civil society contributions to nuclear safeguards

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Abstract:

Civil Society is increasingly involved in the policy area of international arms control. Their opportunities are very limited for compliance control in the nuclear nonproliferation regime due to its particular sensitivity. The severe gaps of nuclear safeguards with respect to the capabilities to detect clandestine facilities render marginal civil society contributions highly influential and controversial. More and more data get available for the civil society that can be used to expose potential violations of the NPT. A systematic framework is presented to classify civil society contributions that allows for a systematic study. This classification uses the two parameters (a) affected safeguards stage and (b) degree of integration with the official procedures. These parameters may have the following defined values:

- *(a) The affected safeguards stage can be*
  - *i. Development and demonstration of new methodologies and technologies*
  - *ii. Fact finding and data gathering*
  - *iii. Sharing and publication of data and information*
  - *iv. Technical analysis of data and information*
  - *v. Determination of non-compliance*
  - *vi. Political interpretation*
- *(b) The degree of integration can be*
  - *i. Without a relation*
  - *ii. Indirect connection*
  - *iii. Informal interaction*
  - *iv. Official contribution or mandate*

A prominent example for civil society contributions is the increasing availability and capability to acquire and analyze satellite images. An emerging field is environmental sampling, analysis and related atmospheric transport simulation. These and other opportunities are put in the systematic framework to discuss their demonstrated and potential impact. In particular, possible contributions that civil society may offer for improving the detectability of unreported facilities and activities are considered with their chances and risks.

Keywords: civil society; verification; nuclear arms control; Nonproliferation Treaty

Summary

Civil society plays an increasing and influential role in many fields of global governance. This is clearly manifested, e.g. in human rights issues, development policy and environmental protection treaties. The international arms control and particularly the verification of treaty compliance are considered as sensitive policy areas for national and international security such that to assign a responsible role to non-state actors is less likely. In principle, self-determined action within the context of the so- called *Whistleblowing* or *Societal Verification* (Rotblat 1993 [1], Deiseroth 2000 [2] and 2008 [3]) is always possible, however, an official agreed-upon role in the official treaty verification is difficult to achieve. Therefore, the contribution of civil society is sometimes called *Unofficial Monitoring* (Bruneau, 2006 [4]). This, however, does not include all other possible cases. Exceptions are found in the conventional arms control. One example is the landmine monitoring operations of the International Coalition to Ban Landmines (ICBL) serving as the main tool to gather information for the Ottawa Anti-Personnel Landmines Convention.

This article focuses on a systematic classification for the various kinds of contribution of the civil society to treaty verification in the nuclear non-proliferation regime. One special motivation for this is deduced from the partial technical failure paired with partial political constraints and interest-driven interpretations of the official system. The glaring gap of the inspection efforts for the Non-Proliferation Treaty (NPT) is the lack of technical and legal instruments to detect clandestine facilities with illegal activities. This remains virulent due to the non-ratification of the additional protocol. Hopes lie in increasing data that are also made available to help expose possible treaty violations. Satellite pictures by Yongbyon, Natanz and Al Kibar are common examples. New efforts are being made to close the identified loopholes of the official verification with new technologies. Based on a systematic classification, this paper analyses systematically which contribution the civil society can offer on different levels of verification and with varying degrees of integration to the official procedure.

1. Objectives and competences of verification

The United Nations[1] officially defines verification of international treaties, as follows:
"Verification involves the collection, collation and analysis of information in order to make a judgement as to whether a party is complying with its obligations."

The verification of a treaty ensures compliance and strengthens the international security. The objectives of verification can be differentiated and are multifaceted:
- Assurance of compliance
- Detection of non-compliance to obligations and security
- Deterrence of treaty violations
- Confidence-building through transparency and openness

Verification is considered as an essential element of all arms control treaties and disarmament agreements. The competence is well defined:[2]
"Verification [...] is conducted by the parties [...] or by an organisation at the request and with the explicit consent of the parties [...]."

According to this, only state parties as well as their respective assigned agencies may conduct verification. In the case of nuclear non-proliferation, these are international organisations supported by the states.

This paper investigates the contributions of civil society to the verification of nuclear arms control treaties. The article follows the comprehension of civil society proposed in the work of Gunnar Jeremias (2011 [7]). This is based on the definition of civil society as the "third sector" in distinction from the state and market sectors.[3] Civil society organisations and individuals which are active in

[1] UN General Assembly 2007. [5]
[2] UN Disarmament Commission 1988 [6].
[3] cf Frantz/Martens, 2006: 18. [8]

verification and monitoring generally act independently and regardless of state directives or profit interests. It includes non-governmental organisations, media representatives, technical experts as well as persons who have access to information about illegal activities.

In view of the much-needed expertise, many of these organisations and individuals become almost mandatory members of the academia, and can be described as *epistemic community* according to Haas (1992 [9]). This concept by Jeremias (2011 [7]) also implies, in accordance with the methodology of peace research, the existence of implicit normative commitment of scientists. In this sense, civil society organisations and individual actors fulfil the function of establishing public transparency in compliance issues.

Representatives of civil society cannot take verification functions directly. However, their expertise could be considered by states or other competent international organizations under certain conditions.

2. Systematic examination of the contribution of the civil society to verification

Generally, it can be declared that the information acquired and reported by the civil society can be used by states and the IAEA to stimulate, compare or supplement their verification activities. Even more substantial is definitely the added input value of civil society, thereby increasing the transparency of databases and improves the findings of official verification as well as the public dissemination of information about suspected illegal or doubtful activities.

To better estimate the contribution of the civil society to verification, this article suggests a systematic analytical framework to classify concrete activities into different levels of verification and determine how strong they are integrated into the official verification process. These two parameters define a matrix in which each civil society contribution can be classified. The columns cover the level and the rows the degree of integration (see table).

The levels of verification include three stages (fact-finding, review, assessment), which are designated by Dekker (2001 [12]). They are expanded with the level “Determination of non-compliance through legal and normative interpretation” and framed by the preparation-phase and post-processing-phase. The degree of interaction, proposed by Meier/Tenner (2001 [13]) is added to the case that a norm does exist but not a treaty with official verification.

The “political assessment of non-compliance” (which is hereby referred to as post-processing phase) is not included in the area of verification but rather in the area of compliance policy. The dividing line between verification and compliance is patterned after the work of Gunnar Jeremias (2011 [7]). He recognized that the verification debate of the 80s was highly influenced scientifically leading to a positivist perspective on compliance. However, in many cases, this is not compatible with the reality of the treaty or with state compliance policies. Since treaty norms are subject to interpretation, the evaluation of compliance is always a political action, which is seen as subsequent step to the actual verification based on the used definition. With this perception, Jeremias [7] confirms the findings of Chayes and Chayes (1995 [14]) that compliance assessment is always a political process.

Each contribution by the civil society can be classified in the matrix including the capabilities and competencies of different actors. Civil society actors contribute, for instance, not only in the disclosure and dissemination of information but also in political assessments. The better they can achieve this task, the more access they have to restricted information. Independent experts with special understanding of data and analytical procedures are also capable to conduct technical analysis and legal examinations. They can also develop and test novel methods. The prerequisite for the success of these contributions is the freedom of expression and legal protection from disseminating information, which is undesirable by the state.

The presented analysis matrix here is only a rough example. A systematic survey of a great number of historic activities by the civil society requires further research. However, with the following list, a preliminary evaluation can be made on the positive effects of civil society activities for treaty verification. Each of these will be illustrated by a practical example.

- Important contributions to the development and demonstration of novel verification methods have been achieved. Thus, the foundations of new verification measures and successful treaty negotiations have been laid. Examples are the development of the ultra-trace analysis with an atom trap for krypton-85 as well as the development and demonstration of seismic detection of nuclear tests.
- The disclosure of problematic activities through whistleblowing facilitates an early warning. Mitchell (2000 [15]) regards the essential contribution of non-traditional sources as fire alarm. The best known example is Mordechai Vanunu and secondly, the Iraqi defector Khidir Hamza.
- Open access technologies enable civil society actors to verify nuclear activities. This happens also at instances where the official verification system fails. Novel sensors provide additional opportunities. Relevant examples are found in the analysis of satellite images for the detection of nuclear facilities. The surface nuclear tests were detected worldwide by many radiation protection laboratories and were reported in many scientific publications.
- Similarly, the civil society can provide additional information to fill up the gaps of verification and make recommendations for political decisions. This is mostly indirectly facilitated through open source information. Satellite images are often provided on the internet, analysed and annotated after a location has been detected via other means, like the assumed Syrian nuclear reactor after the Israeli attack.
- Confidential treaty documents are purposely forwarded to non-state actors who do not hesitate to publish them via internet. The goal of transparency, which is only marginally supported by the official system, can then be achieved through this. The website WikiLeaks is particularly popular in this matter. For years, the confidential reports on the Iranian nuclear program are published immediately after their completion through the IAEA on the website of ISIS (Institute for Science and International Security).
- The civil society also has a function in the processing and presentation of open knowledge base. This is important most of all for countries without own NTMs to lessen knowledge gaps. They help achieve transparency for public information and in emergency cases, compose counterfactual statements against official declarations of single states or verification organisations. The database on seismic events of the U.S. Geological Survey (USGS) is one example. The location, point in time and the seismic magnitude of the second North Korean nuclear test was published by Kalinowski (2009a [17]) via press release and finally transmitted by news agencies in ten different languages.

This list could be supplemented by theoretically possible activities of the civil society. However, this article does not attempt to provide a more speculative view.

However, there is also the risk of negative impacts. Groups of the civil society can be prone to an interest-driven focus of work. Normally, the data used by these groups is not authenticated and thus, quality control is only possible to a certain extent. There is a potential risk of false alarms which are difficult to verify and the risk of false accusations motivated by unilateral interests.[4] These risks have to be counteracted and addressed without restraining the positive effects.

The civil society can strengthen at least partially the verification of nuclear arms control treaties. It can contribute to achieve the different goals of verification (assurance of compliance, detection of non-compliance, deterrence of violation of the treaty and transparency) more completely. The potential of the civil society to contribute to the verification can still be further improved and utilized. Proposals for the improvement of quality and scope of civil society contributions especially for their formal integration into the official process of verification can be found in Crowley/Persbo 2006 [16]. Deiseroth 2008 [3] demands a more effective legal protection for individuals exercising their responsibility. This paper works on the thesis that the role of non-state actors in the verification of nuclear arms control treaties can be strengthened with the support and use of novel scientific and technological developments and methods.

[4] Mitchell 2000 [15] discusses the financial costs of the analysis of false alarms and their political costs

3. Acknowledgement

Special thanks go to the DSF (Deutsche Stiftung Friedensforschung) for the financing of this work. Thanks to the members of the research group on verification and monitoring of international treaties of the Carl Friedrich von Weizsäcker - Centre for Science and Peace Research (ZNF) for the inspiring discussions and insight. I thank Julie Ann Daludado for translation of the German manuscript and Paul Kaiser for editorial support.

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Table 1: Classification of civil society actors in regard of the affected safeguard stage and the degree of integration with official procedures

Affected safeguard stage Degree of integration with official procedures	Development and demonstration of new methodologies and technologies (preparation phase)	Fact finding and data gathering	Sharing and publication of data and information	Technical analysis of data and information	Determination of non-compliance	Political interpretation (post-processing phase)
Without relation (e.g. treaty or verification is non-existent but a norm)	Specialised scientist	Whistleblower (e.g. M. Vanunu)		e.g. ZNF-analysis of the North Korean nuclear tests		Experts for the nuclear weapons control regime, Technical specialised non-governmental organisations
		e.g. seismic stations	e.g. seismic networks	e.g. seismologists	Arms-control specialists specialised think tanks and networks	
Indirect connection (e.g. distributed via open source)	Scientific publications Commercial available technologies	Whistleblower		e.g. ISIS satellite images e.g. radionuclide experts		
		e.g. commercial images e.g. radiation protection laboratories	Illegal information transfer (e.g. Wikileaks.org) Non-governmental organisations with access to information e.g. NCRI			
Informal interaction	Specialised associations and networks e.g. iGSE, ESARDA, INMM	Defectors		?	technical advice	Lobbying and technical advice
		Potentially NCRI as well				
Official contribution or mandate	e.g. GSE, ISS, national support programs for the IAEA	non existent	Non existent	Non existent	Non existent	Non existent

Setting-up the Nuclear Security Training Centre

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Abstract:

The Joint Research Centre, based on its experience and expertise in nuclear security, is involved in the set-up of the European Nuclear Security Training Centre that will address all aspects from detection to response to cases of illicit trafficking of nuclear and radioactive materials.

The original concept foresaw the establishment of the training centre at two locations based on the specific competences of the involved JRC Institutes, the Institute for the Protection and Security of the Citizen (IPSC) in Ispra, Italy and the Institute for Transuranium elements (ITU) in Karlsruhe, Germany. IPSC would be in charge of the part dedicated to the detection and the local response and ITU of the response and nuclear forensics

The Nuclear Security Unit of IPSC has set-up the appropriate infrastructure and developed peer reviewed training syllabi with the support of its international partners in the field for, on the one hand, front line officers and , on the other hand, future national trainers. Several pilot training sessions were organized for both types of audiences in 2009 and 2010.

This paper reports on the experience gained during that initial phase that will be useful for the transfer of the totality of these training activities to Karlsruhe which is planned this year.

Keyword: Training, Nuclear Security, illicit traficking

1. Introduction

The international community concerned by the non-proliferation and the nuclear security has set-up large support programs that include the deployment of detection equipment worldwide. These modern systems are usually run by officers that are not specialists in the detection of Nuclear and other Radioactive Materials (NRM). The training of the personnel in charge is thus of utmost importance to efficiently counteract and interdict the illicit trafficking of NRM.

The Joint Research Centre has a long experience and proven expertise in the field and participates to the international efforts by implementing corresponding support programs on behalf of the European Commission. As part of the projects dealing with border monitoring activities, training is a key component. The JRC has therefore developed the corresponding capability and a facility dedicated to the training on detection and response to illicit transport of NRM has been set-up at the JRC Ispra.

This paper presents the Security Training Centre (SeTraC) facility and reports on the activities that occurred since its first session in September 2009.

2. Set-up of the Nuclear Security Training Centre at Ispra: SeTraC

2.1 Concept

The Nuclear Security Training Centre has been designed since its origin to combine necessary lectures with hands-on training. The concept of a good balance between theoretical classes and practical exercises using real nuclear and radioactive materials has driven the establishment of the SeTraC.
The target audience, front line officers and their trainers, has impacted as well on the construction of the Centre.
Therefore, the concept included outdoor facilities for in-field exercises as close as possible to real conditions, laboratories for practical and demonstrations and a classroom with the necessary modern media equipments.

The development of the training centre benefited of the large support of the US Department of Energy, National Nuclear Security Administration (DoE/NNSA) that invited JRC experts to visit their HAMMER Training facility located in Richland (Washington State). JRC attended to the first day of a training session for Colombia Customs Officers, visited the premises and exercise fields and took benefit of the US experience in the field shared in a very open and collaborative manner that speeded up the establishment of the EU Centre. This support has continued during the implementation phase and will be described in the paragraphs below.

2.2 Infrastructure

The HAMMER training facility is a huge training centre addressing a large panel of security topics. The concept of the SeTraC facility intended to reproduce the part dedicated to nuclear security directed to Front Line Officers and their trainers.
Based on this input and with the initial support of the National Nuclear Security Administration, the initial design included dedicated classroom for theoretical lectures, laboratories for practical and outdoor hands-on training facility mimicking as close as possible the environment of the trainees in their actual daily duties.

Two classrooms are available at the SeTraC. The smaller one can host up to 8 participants and is located close to the outdoor facility. This room also hosts the Central Alarm Station of the Radiation Portal Monitor (RPM). The second, larger, can host 20 trainees and provides the internet access used for the Training of Trainers type of courses. Both classrooms are equipped with modern media means allowing all kind of lectures (presentations, audio and video…). A big room with simultaneous translation means is available at Ispra site. Figure 1 shows the set-up of the smaller classroom.

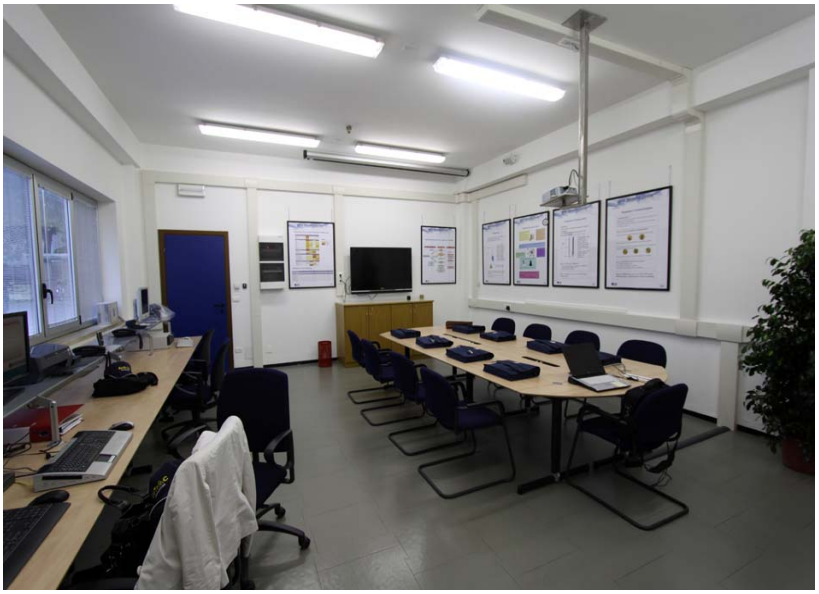

Fig.1: SeTraC smaller classroom and the Central Alarm Station.

The curriculum that has been developed includes exercises on the principle of detection of radioactive and nuclear materials. Parameters such as distance, shielding and exposure time are demonstrated to the students during practical in small groups. These exercises take place in dedicated laboratories hosted in the building of the former Ispra ESSOR research reactor. The room includes separate locations to host 4 groups at the same time, the necessary radioactive sources (thus the detection of the different type of radiation (alpha, beta, gamma and neutrons) can be demonstrated) and detection equipment.

Outside the building and within the nuclear island that offers the mandatory physical protection, space allowing the use of vehicles has been made available for hands-on exercises. In case of adverse weather conditions, a covered place is available to conduct search exercises in vehicles.
Fixed detection equipment for pedestrian (inside the classroom) and for vehicles (outside) is available for practical exercises. The latter has been donated by the US NNSA in the frame of the support to the establishment of the SeTraC (see figure 2).

Fig. 2: Outdoor training area – the vehicle Radiation Portal Monitor.

2.3 Training courses

The Border Monitoring Working Group (BMWG), established under the auspice of the IAEA and coordinating the nuclear security activities of the major international donors (US Second Line of Defence program, International Atomic energy Agency, European Council and European Commission), has set-up a technical working group for the creation of a common syllabus to train the front Line Officers. Participants from the IAEA, SLD and the JRC met in Ispra to define the agenda of a typical one-week training session and gathered the available material from the three organisations. The corresponding syllabus has been jointly reviewed and agreed after the necessary iterations and published as a BMWG product. The agenda of the course is given in Annex 1. The pilot training session, that inaugurated the SeTraC facility, took place in 2009.

Based on the successful experience and taking into account the demand for an advanced course for the trainers of the beneficiary countries, the same group created in 2010 a "Train of Trainers" syllabus. This kind of training aims at building capacity in the beneficiary country to provide the basic training to Front Line Officers. Such capacity allows addressing a large number of concerned staff, training new personnel and refreshing the knowledge of others. Besides the theoretical and hands-on sessions, the participants are requested to prepare presentations concerning technical and / or legal matters to develop their didactic skills. The first pilot session was organised in 2010 and lasted two weeks (8 days). Based on the feedback of this first course, the session was reduced to one week, condensing

the primary syllabus accordingly. The agendas of both the pilot course and the final agreed version are provided in Annex 2.

3. Operational feedback

3.1 Pilot training sessions

The establishment of the Nuclear Security training Centre at the JRC Ispra (JRC) is part of the recommendation of the EU CBRN Action Plan and as such was partially financed by DG JLS (Justice, Liberty and Security - now DG HOME). The Administrative Arrangement signed between JLS and the JRC included the provision of a pilot training session that took place in September 2009.
In close collaboration with SLD, 8 Security and Police Officers from the Shannon Airport in Ireland were selected to follow the first training course. Under a US support program, the airport has been equipped with detection systems to check international flights to the United States during their stop-over. This procedure qualifies the controlled flights as domestic ones.
Corresponding detection of radioactive and nuclear materials was demonstrated and the procedure to properly respond explained during the pilot session. Exercises with hidden real materials in vehicles and luggage trained the participants to react to situation similar to what could be faced during their daily duties. Officers were familiarized with Radiation Portal Monitors, Personal Radiation Detectors, Radioisotope Identifier Devices, safe search procedure and reporting, profile analysis and different types of alarms.

The second pilot training session took place in June 2010. Trainers from Asian countries (Indonesia, Malaysia, Pakistan, Philippines) participated to an 8 days training course with IAEA support. This "Train of trainers" course built upon the syllabus for Front Line Officers with a more detailed series of technical lectures as well as specific courses and home exercises to develop training skills. The feedback from this first experience led to jointly decide with our partners (IAEA and SLD) that a one week course will be more convenient. Accordingly, the agenda was reshuffled and adapted to the new timing which is now the normal duration for this kind of training.

3.2 Training courses

Since the first course, the SeTraC has organised training sessions for both the EC as well as for external customers upon request. The two types of courses have been held in 2010 and are summarized below. The increasing number of sessions corresponds to the initial plan for establishing a sustainable nuclear security training activity at JRC.

- Officers from the Lebanese Customs participated to a one week training course in October 2009 under IAEA support.
- Front Line Officers from Irak were trained in November 2010; the course was organised by the IAEA and hosted by the JRC. During the same month, JRC trained Croatian Officers in the frame of the enlargement process. This last session gave the opportunity to test a training course provided into the mother tongue of the trainees: the syllabus was translated into Croatian, simultaneous translations from English to Croatian was delivered during the lectures and Croatian interpreters helped during the practical exercises.

- Finland has set-up a national training plan and relies on the JRC facility for the practical part of the training curriculum. After the theoretical session organised in Finland with the presence of the JRC, the first practical session was hosted in April 2010 at the SeTraC where the JRC team organised jointly the three days training. Feedback from the session has been taken into account and the course adapted accordingly as this support is planned to be provided annually.

- One more session was organised for the trainers under IAEA auspices for African countries (Namibia, Ghana, Tanzania, and Uganda). The one week training course took place in September 2010.

- As a spin off of the SeTrac activity, the JRC has been invited to participate also in in-situ training courses under IAEA request.

4. Future of the Nuclear Security Training Centre

On January 1st, 2011 the Nuclear Security Unit shifted from the Institute for the Protection and Security of the Citizen to the Institute for Transuranium Elements (ITU) located in Karlsruhe. The Ispra site decommissioning program and the transfer of nuclear material and radioactive sources to ITU impacted on the development of the SeTraC project and a new strategy was adopted.
The SeTraC facility will be merged with the training centre dedicated to response and nuclear forensics being established in parallel at ITU. A larger concept, the EURATOM school, grouping all JRC nuclear education and training activities will be implemented in the coming months. The EURATOM school will therefore include all nuclear security training activities currently provided at JRC Ispra. A dedicated facility called EUSECTRA is being built and should be operational by the end of 2012.

In order to assure the continuity for the customers, the SeTraC will organise five training sessions in 2011 upon request. Four "Training of Trainers" will be sponsored by the IAEA while 1 "Front Line Officers" will be provided for SLD.

5. Conclusion

The establishment of the Nuclear Security Training Centre at JRC Ispra in 2009 included the design of the facility and the development of the necessary syllabi. The Nuclear Security Unit has benefited from the experience and support of the Members of the Border Monitoring Working Group for an integrated approach in line with the international activities in the field.

The SeTraC has been set-up during the first semester of 2009 and the first pilot training session took place in September of the same year. Training courses for Front Line officers and their Trainers are routinely provided since then for the European Commission as well as for external customers. Nuclear training and education activities will be part of the EURATOM school in the near future and therefore the SeTraC activities will be transferred by the end of next year to the Institute for Transuranium Elements in Karlsruhe, Germany.

The experience gained will be used for the transfer/integration of the SeTraC within the EUSECTRA facility in the frame of the creation of the EURATOM school atJRC/ ITU.

6. Annexes

6.1 Training for Front Line Officers

Monday	Tuesday	Wednesday	Thursday	Friday
9:00 - 9:30 Opening Session Address by: SLD (E. Melamed) JRC (P. Frigola)	9:00 - 9:45 Overview of Equipment (P. Peerani)	9:00 - 9:20 How to document an Alarm (V. Rouillet Chatelus - IAEA)	9:00 - 9:30 Analysis of Real Cases (J. Bagi)	9:00 - 9:30 Reachback in Nuclear Security Context (P. Daures)
13:30 - 14:15 Basic Radiation Physics (P. Richir)	9:45 - 10:15 Model Action Plan (V. Rouillet Chatelus - IAEA)	9:20 - 10:15 Radiation Portal Monitors (V. Berthou)	9:30 - 10:15 B&C Proliferation & Terrorism (L. Olmedo - CEA/DAM)	9:30 - 10:15 Presentation of the National Situation (J. Francis - Irish Authorities)
10:15 - 10:30 Coffee Break	10:15 - 10:30 Coffee Break	10:15 - 10:30 Coffee Break	10:15 - 10:30 Coffee Break	10:15 - 10:30 Coffee Break
14:15 - 15:00 Radiation protection (D. Giuffrida)	10:30 - 11:15 Operational Response to Detection (T. Pelletier - CEA/DAM)	10:30 - 11:15 Illicit Trafficking Database (V. Rouillet Chatelus - IAEA)	10:30 - 11:15 Crime Scene Management (M. Wallenius)	10:30 - 12:00 Table Top exercise (JRC/SLD)
11:15 - 12:00 Introduction to WMD and Threat Awareness (P. Peerani)	11:15 - 12:00 Operational Response to Detection (Demonstration) (T. Pelletier - CEA/DAM)	11:15 - 12:00 Profile Game (D. Pappas - SLD)	11:15 - 12:00 Nuclear Forensics (M. Wallenius)	
12:00 - 13:30 Lunch Break	12:00 - 13:30 Lunch Break	12:00 - 13:30 Lunch Break	12:00 - 13:30 Lunch Break	12:00 - 13:30 Lunch Break
15:15 - 16:00 Practical Demo on Radiation Physics PERLA (M. Marin Ferrer, J. Bagi)	13:30 - 14:00 Handheld Equipment Specific Presentation (D. Pappas - SLD)	11:30 - 14:00 RPM Specific presentation (D. Pappas - SLD)	13:30 - 15:00 Identification of Radioactive Sources and Nuclear Materials PERLA (M. Marin Ferrer, J. Bagi, H. Tagziria)	13:30 - 14:30 Discussion and Course Feedback
9:30 - 10:15 International Legal Instruments for Nuclear Security (V. Berthou)	14:00 - 16:00 Hands-on Training on Secondary Inspection (JRC / SLD)	14:00 - 16:00 Hands-on Training on Portal Monitors (JRC / SLD)		14:30 - 15:00 Award of Certificates
15:00 - 15:15 Coffee Break			15:00 - 15:15 Coffee Break	
10:30 - 11:15 International Transport Regulation (T. Pelletier - CEA/DAM)			15:15 - 16:00 Radioactive Sources Database and Visual Recognition (M. Marin Ferrer)	
16:00 - 16:30 Debriefing and Q&A (All lecturers)	16:00 - 16:30 Debriefing and Q&A (All lecturers)	16:00 - 16:30 Debriefing and Q&A (All lecturers)	16:00 - 16:30 Debriefing and Q&A (All lecturers)	

6.2 Training of Trainers

Time	Day 1	Time	Day 2	Time	Day3	Time	Day4
9:00 - 9:30	Registration	9:00 - 9:45	Basic Radiation Physics & Radiation Protection P. Richir	9:00 - 10:00	Legal Framework for Nuclear Security (participants presentations and discussion)	9:00 - 9:30	Authorized Activities involving Nuclear and other Radioactive Material A. Bacheller
9:30 - 9:45	Opening Session (welcome, administrative info) JRC, SLD, IAEA	9:45 - 10:00	Coffee break	10:00 - 10:15	Coffee break	9:30 - 10:00	Export Control and Nuclear Security F. Sevini
9:45 - 10:00	Introduction of Participants	10:00 - 12:30	Demonstration on Radiation Physics I. Bagi	10:15 - 11:15	Principle Radiation Detection V. Berthou	10:00 - 10:15	Coffee break
10:00 - 10:30	Participants' Expectations D. Pappas	12:30 - 14:00	Lunch break	11:15 - 12:45	Overview on Nuclear Security Instruments I. Bagi	10:15 - 10:45	International Catalogue of Sealed Radioactive Sources and Devices A. Bacheller
10:30 - 10:45	Coffee break	14:00 - 16:45	Legal Framework for Nuclear Security (introduction and group work) A. Bacheller	12:45 - 14:00	Lunch break	10:45 - 11:15	International Catalogue of Sealed Radioactive Sources and Devices (exercise on visual recognition) A. Bacheller
10:45 - 11:10	Objectives of the Training & Knowledge Questionnaire A. Bacheller & D. Pappas	16:45 - 17:00	Wrap-up	14:00 - 17:30	Specific equipment presentation (participants)	12:45 - 14:00	Lunch break
11:10 - 12:10	Trafficking in Nuclear and other Radioactive Material P. Peerani			17:30 - 17:45	Wrap-up	14:00 - 15:30	Scenario Development P. Peerani
12:10 - 14:00	Lunch break					15:15 - 15:30	Coffee break
14:00 - 14:45	Training Aids D. Pappas					15:30 - 17:00	Scenario Development (cont'd) P. Peerani
14:45 -15:30	Training & Instruction M. Clarke					17:00 - 17:55	Wrap-up
15:30 - 15:45	Coffee break						
15:45 - 16:45	Selected Teaching Methods M. Clarke						
16:45 - 17:00	Wrap-up						

Time	Day 5	Time	Day 6	Time	Day 7	Time	Day 8
9:00 - 12:30	**Hands-on training on scenarios**	9:00 - 10:30	**Discussion on Scenarios Hands-on Training P. Peerani**	9:00 - 09:30	**Course Preparation M. Clarke**	9:00 - 9:45	**Participants' expectations & Knowledge Questionnaire S. Goheen**
12:30 - 14:00	**Lunch break**	10:30 - 11:00	**Illicit Trafficking Database Programme A. Bacheller**	9:30 - 10:00	**Training Evaluation M. Clarke**	9:45 - 10:00	**Coffee break**
14:00 - 14:45	**Transport Regulation Pelletier**	10:30 - 10:45	**Coffee break**	10:00 - 11:30	**Quiz Games SLD**	10:00 - 11:30	**(Distribution of training & teaching material, training evaluation, concluding remarks) JRC, SLD, IAEA**
14:45 - 15:30	**Operational Response to Detection Pelletier**	10:45 - 11:30	**Analysis of Real Cases I. Bagi**	11:30 - 11:45	**Coffee break**	11:30 - 12:00	**Award of Certificates**
15:30 -15:45	**Coffe break**	11:30 - 12:45	**Case Study SLD**	11:45 - 12:15	**National Training Capabilities (preparation of presenations)**	12:00	**Lunch**
15:45 - 17:15	**Operational response to detection (practical exercise) Pelletier**	12:45 - 14:00	**Lunch break**	12:15 - 14:00	**Lunch break**		
17:15 - 17:30	**Wrap-up**	14:00 - 16:15	**Case Study (Cont'd) SLD**	14:00 - 14:30	**National Training Capabilities (preparation of presenations - cont'd)**		
11:00 - 12:30	**Case Study (Cont'd) SLD**	16:15 - 17:00	**Introduction to Nuclear Forensics - Identifying the Origins of Nuclear & Radioactive Materials M. Wallenius**	14:30 - 16:30	**Presentations and Discussions on National Training Capabilities**		
		17:00 - 17:15	**Wrap-up**	16:30 - 16:45	**Wrap-up**		

Monday	Tuesday	Wednesday	Thursday	Friday
13 September 2009	14 September 2009	15 September 2009	16 September 2009	17 September 2009
9:00 - 9:30 Opening Session	9:00 - 9:30 Principle of Radiation Detection (P. Peerani)	9:00 - 10:15 Group Presentation on Equipment (15 min per Group)	9:00 - 10:15 Hands-on Training on Scenarios (V. Forcina)	9:00 - 10:00 Table Top Exercice (P. Daures)
9:30 - 10:15 Introduction of participants and expectations	9:30 - 10:15 Overview of Nuclear Security Instruments (P. Peerani)			10:00 - 10:15 Documenting an Alarm (P. Peerani)
10:15 - 10:30 Coffee Break	10:15 - 10:30 Coffee Break	10:15 - 10:30 Coffee Break	10:15 - 10:30 Coffee Break	10:15 - 10:30 Coffee Break
10:30 - 11:00 Trafficking in Nuclear and other Radioactive Materials (V. Rouillet Chatelus)	10:30 - 12:00 Operational Response to Detection (T. Pelletier - CEA/DAM)	10:30 - 11:00 Illicit Trafficking Database (P. Daures)	10:30 - 12:00 Hands-on Training on Scenarios (Cont'd)	10:30 - 11:00 Quizz Game (P. Richir)
11:00 - 12:00 Basic Radiation Physics and Radiation Protection (P. Richir)		11:00 - 11:30 Authorized Use of Nuclear and Radioactive Materials (P. Peerani)		11:00 - 12:00 Course Feedback, Assessment and Evaluation by Participants
		11:30 - 12:00 Catalogue of Radioactive Sources (P. Daures)		
12:00 - 13:30 Lunch Break	12:00 - 13:30 Lunch Break	12:00 - 13:30 Lunch Break	12:00 - 13:30 Lunch Break	12:00 - 14:00 Lunch Break
13:30 - 15:15 Practical Demo on Radiation Physics (J. Bagi)	13:30 - 14:15 Operational Response to Detection (Demonstration) (T. Pelletier - CEA/DAM)	13:30 - 14:00 Export Control (F. Sevini)	13:30 - 14:30 Preparation of Participants Presentation on National Training Capabilities	14:00 - 15:00 Award of Certificates and Closing Session
	14:15 -15:00 Legal framework: Group Presentation (10 minutes each)	14:00 - 15:00 Nuclear Forensics (K. Mayer)	14:30 - 15:30 Presentation of Participants on National Training Capabilities (10 min each)	
15:15 - 15:30 Coffee Break	15:00 - 15:15 Coffee Break	15:00 - 15:15 Coffee Break	15:30 - 15:45 Coffee Break	
15:30 - 16:30 National Legal Framework (Introduction and group work)	15:15 - 15:45 International Transport Regulation (Film)	15:15 - 16:00 Analysis of Real Cases (J. Bagi)	15:45 - 17:00 Training Aids and Selected Training Methods (V. Berthou)	
16:30 - 17:00 Course Preparation (V. Rouillet chatelus)	15:45 - 17:00 Preparation of Participants Presentation on Specific Equipment Presentation (Group Work)	16:00 - 17:00 Scenario Development (Group work)		
		Hospitality Event		

Assessing and Promoting the Level of Safeguards Culture in Hungarian Nuclear Facilities

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Abstract:

The Hungarian SSAC has just introduced a comprehensive domestic safeguards verification system consisting of regular comprehensive SSAC verifications in the whole lifetime of the facilities. The main goals of the comprehensive verification system is: (i) to assess the facility's safeguards system compliance with the relevant national legislation and recommendations, (ii) to assess the activities of the facility aimed at maintaining and further developing its safeguards system and (iii) to revise validity of data and information previously provided by the facility subject to safeguards licensing procedures. The maintenance level of the system as well as the available knowledge on the possible needs for change reflect the top management's awareness of this issue and is a good indicator of the present and future effectiveness of the facility level safeguards system and the level of safeguards culture. The structure, preparation, conduction, documentation and initial experiences of the comprehensive safeguards verification system is introduced in the paper.

Keywords: national safeguards system; comprehensive domestic safeguards verification system; safeguards culture; SSAC

1. Introduction

The effectiveness and efficiency of an SSAC greatly depends on how the management in the nuclear facilities is committed to the non-proliferation objectives of the country.

In Hungary safeguards licensing procedures are obligatory to possess nuclear material, launch any activity related thereto, launch any modification important to safeguards, transport nuclear materials, as well as to terminate safeguards requirements in case of terminating nuclear activities. In addition to it, facilities are obliged to maintain a facility level nuclear material accountancy system and create the required conditions for international, regional and national verification activities. It is, however, essential that the above obligations be integral parts of a coherent facility management policy.

Based on very promising experiences in the field of nuclear safety, the Hungarian SSAC has just introduced a comprehensive domestic safeguards verification system consisting of regular comprehensive SSAC verifications in the whole lifetime of the facilities.

The structure, preparation, conduction, documentation and initial experiences of the comprehensive safeguards verification system is introduced below.

2. The comprehensive domestic safeguards verification system (CDSVS)

The introduction of the comprehensive domestic safeguards verification system (CDSVS) by the Hungarian SSAC started with laying down the procedure of the CDSVS in the Hungarian Atomic Energy Authority's (HAEA) Quality Assurance System. The QA procedure for the CDSVS was approved by the General Deputy Director General of the HAEA. Carrying out CDSV falls into the competence of the Department of Nuclear and Radioactive Materials of the HAEA (hereinafter referred to as the Safeguards Department).

2.1. Goal of the CDSVS

The main goal for the CDSVS was defined as follows: to review whether the facility level safeguards system of the organization is run in compliance with the relevant legal instruments and recommendations in force. To reach this goal two tools are to be applied:

a.) to review all the safeguards relevant procedures of the organization. In this review the focus is to check whether procedures for fulfilling the obligations are regulated and to find practical examples for the procedures by the competent staff.

b.) to assess the activities of the organization in view whether it ensures sustainability and improvement of the safeguards system in all levels of organisation, with special regards to the commitment on management level.

The Safeguards Department of the HAEA plans to carry out one comprehensive verification inspection in one of the Hungarian nuclear facilities per year. In 2011, for the first time, the Modular Vault Dry Storage (MVDS) of the Spent Fuel Assemblies was selected for CDSVS. Verification of the management systems (highest management and safeguards division management) as well as safeguards relevant areas as operation and maintenance, accountancy and data provision were selected for verification.

2.2. Verification levels

2.2.1. 'Level – A' verification

As the primary goal of the verification is to assess the commitment of the highest management, verification 'Level A' was assigned to the top management of the organisation. 'Level – A' verification was planned to assess the commitments of the managers in the field of safeguards and the guarantees provided by the management to enable the organization to meet its safeguards obligations.

A list of issues in 6 themes was provided in advance for the management to help preparation for the on site inspection. Issues were grouped in 10 themes. Short description of the issues:

1) External influence (e.g. dependence of meeting their safeguards obligation on political changes, TSOs; public acceptance of their mission, safeguards in their external communication; possible responds of the organization in case of negative effects.)
2) Objectives and strategies (objectives of non-proliferation relevance, consultation process in drafting strategies, possible future plans on any changes in this field)
3) Management functions and their review (selection criteria in the management, evaluation of proper and improper safeguards related decisions, competences, etc.)
4) Allocation of resources (corporate procurement and/or restructuring with non-proliferation and safeguards aspects)
5) Human resource management (reduction of staff - giving priority to safeguards staff; vacancy and fluctuation in safeguards staff; promotion, reward system for safeguards staff, etc.)
6) Training (professional training possibilities for the safeguards staff, safeguards for the staff in general, etc.)
7) Knowledge management (ensuring continuity of safeguards staff, communication channels for safeguards knowledge, etc.)
8) Regulation (regulation work processes in view with safeguards obligations, inclusion of safeguards aspects in revision of documents, etc.)
9) Organization culture (evaluation of the performance safeguards related tasks on individuals' appraisal or on organization's level, who performs the appraisal of the individual in the safeguards unit, etc.)
10) Communication (channels of information from external source to the safeguards staff and vice-versa.)

2.2.2. ‘Level – B verification’

‘Level - B’ was assigned to different safeguards related fields with the following subdivision:

B1 – Safeguards division (analyses of the safeguards division structure, its relation with the highest managements, scope of competences; education background and professional training of the safeguards staff; adequate human resource for the related tasks, etc.)

B2 – Operation and maintenance (availability, authentication and maintenance of the measurement equipment to support the accountancy, measures to ensure safe and secure operation of the safeguards containment and surveillance systems, utilization of the organization’s own operational experience as well as safeguards experience and research and development activities of other organizations; procedures established to enable national and international inspections, e.g. ground pass systems, safeguards duty system with telephone contact availability, etc.

B3 – Accountancy and data provision (internal procedures regulating the nuclear material accountancy and safeguards related data provision system, operation and reliability of the computer based accountancy system, etc.)

2.3. Schedule of the verification

The CDSV is carried out along the following schedule:

1.) Preparatory phase (review and process of the related internal documents of the organization)
2.) On site inspection
3.) Assessment

2.3.1. The preparatory phase

The preparatory phase is very important part of the verification. The Safeguards Department held an initial meeting to prepare the verification. On this meeting goals of the CDSVS and levels of verification were explained to the representatives of the MVDS. Participants of the meeting agreed on collecting the internal documents regulating the tasks of the organization and allocating the responsibilities within the units of the organization. It was agreed that these documents would be provided for the HAEA well in advance of the meeting to enable the staff’s preparation for the verification. Potential participants on the on-site inspection both from the HAEA and the MVDS were discussed but not finalized.

Due to the commitments of top management not subject to their influence, the date originally planned for the inspection had to be postponed. A list of issues for the verification of ’Level – A’ was handed over to the representatives of the meeting to assist them in the preparation of the management for the on site inspection.

In the preparatory phase representatives of HAEA on the on-site inspections will study the internal documents of the MVDS and finalize the list of issues on the areas assigned to them.

2.3.2. The on site inspection phase

The on site inspection is planned to be conducted according to the following agenda:

- Kick-off meeting – information on the goal and areas of inspection, and the methods to be applied
- Inspections to be conducted
    - with participants identified in advance
    - based on list of issues for revision (While level – A list of issues were handed over in advance, list of issues for the level – B areas will be used on the on-site inspection only)
    - detailed records on answers and other observations will be prepared by the inspectors

- Closing meeting – preliminary evaluation will be given. There will be possibility given for the licensee to argue the preliminary evaluation results.

2.3.3 Assessment phase and corrective actions

After the on site inspection, HAEA will finalize the report on the inspection and send it to the MVDS for comments.

The report shall focus on identifying best practices and deficiencies, if any, and clearly state the authority's positions how to make corrective actions.

The MVDS shall comment on the main findings and formulate its position on the HAEA's conclusions and recommendations. In its reply MVDS shall identify the means and timeframe of the corrective actions to be performed.

Taking the MVDS response and proposal full into account, the HAEA will issue a regulatory resolution on the corrective actions to be taken and determine deadlines for each.

In addition the HAEA will establish the next review program of the CDSVS focusing on those areas where corrective actions were identified.

3. Conclusion

Although the new comprehensive domestic safeguards verification system has just been introduced and started and has not been completed yet, the HAEA is confident that this new program will reach the following objectives:

The management will be more aware on its safeguards obligation. 'Level – A' list of issues will help the management to analyse the set of documents of the MVDS, from the organization's strategy documents to the low level internal documents. Safeguards related scope of competence needs to be assessed from the top management level to the safeguards officers' level.

Review all the safeguards relevant procedures of the organization will help to disclose the possible gaps in the regulation of the procedures or in the scope of competence.

The need for sustainability of the safeguards system and improve in performance at all levels within the organization will clearly be highlighted through the whole verification process.

Nuclear safety and security culture are well respected and developed in Hungary. The first CDSVS will contribute to empowering the management on the importance of safeguards. The CDSVS will enhance the commitment of the MVDS top management through a well structured dialogue with the HAEA.

In this way improving the nuclear safeguards culture in the organization is expected to get the same importance as nuclear safety and security culture.

24 Poster session I

The ESARDA Verification Technologies and Methodologies Working Group: Addressing Verification Challenges Globally

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Abstract

*This article relates the history of the creation of the ESARDA working Group on Verification Technologies and Methodologies (VTM) which is closely linked to the Association's one. After a brief introduction to ESARDA, I gives a quick overview of the objectives, activities and product of the VTM as the prospects for the years to come. ESARDA, the European Safeguards Research and Development Association has just passed the age of 40 and could look back on a wealth of achievements in the European safeguards area. ESARDA strives to raise the awareness for the non proliferation both in Europe and on an international scale. One of the youngest Working Group of ESARDA is the VTM Working Group which has been founded in 2003. The VTM approach is to consider verification issues globally "**the big picture**" for a mutual benefit of different regimes. VTM mission stems from the acknowledgment of the need for a forum where to discuss on verification issues of multinational instruments. The topics addressed in VTM cover a wide range of verification issues: nuclear and non-nuclear treaties, research and development of innovative verification technologies, synergies between verification regimes, implementation of advanced verification technologies and approaches as IAEA safeguards, CTBT, exports controls, Chemical and Biological Weapons convention, Arms Control and Disarmament issues, Fissile Material Cut-off treaty, Environmental treaty (Kyoto protocol). The VTM try to provide the non proliferation and security community with expert advice on modern verification technologies and methodologies and to act as a forum for the exchange of relevant information in this area. Individual working group members volunteer to prepare discussion and working papers, subgroups have been established, conferences and meetings with special topics are performed. During the years 2007, 2008 and 2009 the VTM had presentations and discussions on various topics which will be presented as examples Two publications have been issued on treaty verification and satellite imagery during the last years and a further book publication is planned on the complex field of proliferation resistance. The next challenges will be to support the evolution of EURATOM and IAEA safeguards, the strengthening EU security, the verification of FMCT and in a European context the entry into force of the Lisbon Treaty. The paper will present the objectives and the scope and achievements of the VTM and highlight its relation with the non proliferation and security community, IAEA, EURATOM, EU Member States laboratories and institutions, US DOE, and other associations in particular with INMM IS Division and NAC Division.*

Keywords: Verification, Technologies, methodologies, global approach, detection, undeclared activities

I. ESARDA[1]

ESARDA, the European Safeguards Research and Development Association, is comprised of European organisations actively involved in the Research and Development of Nuclear Safeguards. Close contacts are held with international partners and organisations. The control of civil nuclear material is mandatory within the EU territory in line with the Treaty establishing the European Atomic Energy Community (“Euratom Treaty”) which entered into force in 1958. The Treaty on the Non Proliferation of Nuclear Weapons, which entered into force in 1970, provides for Nuclear Safeguards on a world-wide basis. The 1960s were a period of intense R&D activity in safeguards, due to the rapid expansion of nuclear energy production. ESARDA was created in 1969. Current members of

ESARDA are: AREVA (France), ATI (Austria), BNG (UK), CEA (France), CNCAN (Romania), EDF (France), ENEA (Italy), European Commission, FZJ (Germany), HAEA (Hungary), IKI (Hungary), IRSN (France), Ministry of Economy / MITyC (Spain), NRI Rez (CZ), NNL (UK), NRPA (Norway), SCK-CEN (Belgium), Sellafield Ltd (UK), SFOE (Switzerland), Springfields Fuels Ltd (UK), SSM (Sweden), STUK (Finland), UKAEA (UK), VATESI (Lithuania), WKK (Germany). URENCO is candidate. In addition, ESARDA counts three individual members. The European Commission represents the European Atomic Energy Community (EURATOM). Representatives from other organisations that are not member parties, amongst them the IAEA, ABACC and INMM, regularly take part in ESARDA activities. The ESARDA network includes national regulatory authorities (carrying out the controls), operators of nuclear facilities (those being controlled), and research centres and universities (carrying out the safeguards-related R&D).

ESARDA main objective is to assist the European safeguards community with the advancement of safeguards, enhancing the efficiency of systems and measures, as well as investigating how new techniques can be developed and implemented. ESARDA also strives to fulfil an educational role and to reach the general public and, in particular, those interested in safeguards. ESARDA aims to bring together all those involved in safeguards, so that progress and continuous improvement in international safeguards can be achieved efficiently and to a professional standard. The principal issues are co-ordination of research, exchange of information and joint execution of R&D programmes. To this end, the following activities take place:

- **Annual Meetings and Symposia**: These provide an opportunity for collaboration and the exchange of scientific information. The latest developments and different perspectives on tackling the challenges in the field are presented and debated at the meetings, which traditionally take place in May. Alternating with open symposia, a biennial internal meeting takes place where all ESARDA working groups convene and have the opportunity to discuss issues of common interest in joint meetings.
- **Working Groups:** They are established to promote and undertake collaborative R&D and information exchange activities in particular fields. The R&D activities are performed by more than 100 experts (members or observers of ESARDA) collaborating within the various working groups.
- **ESARDA Bulletin and web site**: In the ESARDA Bulletin, scientific and technical articles relating to safeguards and verification are published. The Bulletin includes a dedicated section for peer reviewed papers, in addition to more general information and news on ESARDA and its members. The web site features the latest issues of the Bulletin, as well as scientific and practical information about the working groups and symposia. It also contains general information and reference materials relating to safeguards (Treaties, etc.).

One year after its 40th anniversary, ESARDA can look back on a wealth of achievements in the European safeguards area, to which its members have substantially contributed. Also today, ESARDA is more active then ever, both due to an extended partnership (with many new members joining in recent years) and a pro-active attitude to tackle new and upcoming issues through its many sectoral Working Groups.
ESARDA constitutes today of 9 working groups : Non Destructive Analysis, Destructive Analysis, Containment and Surveillance, Verification Technologies and Methodologies, Integrated Safeguards, Nuclear Material Accounting and Control Audit Focus, Training and Knowledge Management, Fuel Fabrications Plants, Editorial Committee. Most of these working groups organise regular meetings in which they discuss the progress, achievements, problems, challenges and future needs for R&D. They jointly work out solutions to problems and stay abreast of the latest safeguards R&D aspects.

Due to the fact that ESARDA has a healthy mix of members from research, authorities, safeguards inspectorates, industry etc, and that the Working Groups do not work in isolation, ESARDA also discusses and gathers very valuable experience with respect to aspects of safeguards implementation on national and international level.

II. History of the VTM

The need for ESARDA to have a working group addressing the new verification issues which has been raised along the nineties and the new tools to deal with has been pushed by Dr Gotthard Stein backed by some other members of ESARDA. Events like, the IAEA safeguards crisis in Iraq, and

DPRK, the adoption of new safeguards and non proliferation instruments to give the IAEA the means to verify not only the correctness of state's declaration but above all their completeness as the Additional Protocol to the Comprehensive Safeguards agreements the emergence of new technical means as the environmental monitoring, the remote monitoring the open sources survey, to detect undeclared activities the definition of new methodologies to implement the verification, the adoption of new disarmament treaties as the Chemical Weapons Convention, the Comprehensive Test Ban Treaty, the Fissile material Cut off, all the issues related to the former Soviet Union disarmament, the indefinite prorogation of the Non Proliferation Treaty, the responses to give to the threats of the proliferation of Weapons of Mass Destruction and their means of delivery have been the main issues which fuelled the reflexion on the evolution of ESARDA. This illustrate that the rationale for the creation of a Verification Technologies and Methodologies working group is deeply rooted in the history of ESARDA as related by its President Mrs. Elina Martikka in the article issued for the forty years anniversary of the association[2].

At that times, the idea of exploring the benefit of the synergies between other verification regimes oriented to the control and reduction of weapons of mass destruction is an area which has already been tackled by ESARDA in several sessions of symposia, in particular during the ESARDA annual seminar “Modern Verification Regimes : Similarities, Synergies and Challenges” held at Helsinki in May, 1998[3]. Although each verification regime has its own specificity in the implementation its regulations, the general methodologies present similarities and some technologies could be used by different systems; important synergies can therefore be found and exploited. The discussions held during the Helsinki symposia sowed the seeds of the VTM working group. In 2000, pointing out that the international safeguards and non proliferation context has dramatically changed since the last evolution of ESARDA, a new reflexion group has been set up to allow ESARDA to cope with the new issues. The Reflexion Group recommended[4] that ESARDA could give further consideration to the role it can play in helping to keep its members abreast of safeguards- related developments in the wider subject of nuclear non proliferation. For the possible extension of the interest of ESARDA, a clear distinction is to be made between verification regimes dealing with the nuclear area and verification regimes dealing with non-nuclear areas (as chemical and biological).

To meet this recommendation and fulfil the need, **Dr Gotthard Stein** (Forschungszentrum Jülich) created the Verification Technologies and Methodologies working group (VTM WG) in 2002 with the help of some other ESARDA members. He was the first Chairman and gave the WG its specific personality. The VTM WG held his first meeting at JRC/ISPRA in February 2003 to draft its terms of Reference and set up a programme of work and discuss issues of interest.

III. Mission / Objectives: understanding "*the big picture*"

The mission of the ***ESARDA Working Group on Verification Technologies and Methodologies*** stems from the acknowledgment by ESARDA members of the need for a forum where to discuss on verification issues of multinational instruments. The VTM WG aims at providing the safeguards community, elements with expert advice on modern verification technologies and methodologies and to act as a forum for the exchange of relevant information in this area. To this end the VTM Working Group has been charged with the following tasks (figure 1):

- Identify, evaluate and promote verification technologies and methodologies which can be applied in specific verification areas (both nuclear and non-nuclear).
- Assess science and modern technology and identify possible new verification technologies and methodologies.
- Identify and promote research areas for new verification technologies and methodologies and support the creation and realization of research networks.
- Stimulate discussions and communications between different verification institutions in Europe and elsewhere
- Offer a forum for experts in different verification regimes to meet and exchange ideas on a regular basis.
- Encourage the private sector in the development and commercialization of verification technologies and methodologies

IV. Topics addressed

The topics addressed in VTM WG cover a wide range of verification issues on nuclear and non-nuclear regimes treaties, research and development of innovative verification technologies, synergies between verification regimes or the implementation of advanced verification technologies and approaches constitutes the workload of the VTM group and determines the agenda of the meetings.

The VTM WG operates as a transverse Working Group: The Topics which are addressed are strongly linked to those addressed in other ESARDA Working Groups as Destructive Analysis (DA), Non Destructive Analysis (NDA), Confinement/Surveillance (C/S)), Implementation of Safeguards (IS), the Nuclear Material Accountancy and Audit Focus Group (NMACAG). Objectives and topics addressed by the VTM are summarize in the figure1

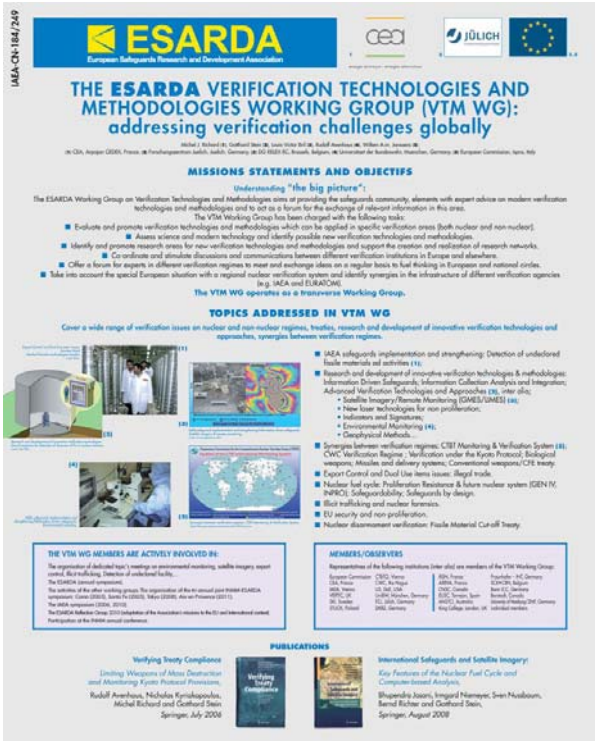


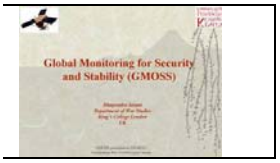
Figure 1 : Poster on the VTM objectives and activities.

Since its inception in 2002, the VTM addressed topics related to safeguards implementation, non proliferation, nuclear security, disarmament and treaty verification, technologies/detection equipment R &D: how to use it?, method of implementation/inspection, verification & monitoring, legal and institutional aspects as illustrated above (inter alia): :

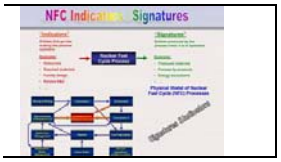
- IAEA safeguards implementation and strengthening
- Research and development of innovative verification technologies
- Synergies between verification regimes: nuclear, Chemical, Biological, Missiles Conventional weapons
- The implementation of advanced verification technologies and approaches
- Chemical Weapons Convention Verification Regime
- Environmental Monitoring: Verification under the Kyoto Protocol
- Environmental Monitoring: environment and wide Area Monitoring
- Comprehensive Test Ban treaty Monitoring & Verification System

- Nuclear Forensics: illicit trafficking/bulk analysis
- EU Security: Non-proliferation of weapons of mass destruction
- Export Control and Dual Use items issues
- Information Driven Safeguards: Role of information collection, analysis and integration for

International Verification



- Satellite Imagery and International Security/Remote Monitoring: GMOSS LIMES & GMES, Seismic Monitoring
- Exploring new laser technologies and laser Measurements for Safeguard
- Proliferation Resistance: future nuclear system GEN IV, INPRO, safeguardability, safeguards by design
- Preventing the Spread of WMD Expertise from Former Soviet Military Scientists: Discussion of ISTC & STCU Issues
- Nuclear disarmament verification: cut-off treaty, excess material disposition, trilateral initiative



- Exploring the potential of Novel Technologies for IAEA Safeguards

V. Highlights on recent years activities

The VTM WG is involved in the participation and the management of meeting in several frameworks:

- First) the VTM WG is actively participating to the life of the Association in particular with paper presentation during the annual conference and symposium, bringing support to the the Steering Committee and the Editorial Committee

In particular, the VTM is fully involved along with the other WGs and the ESARDA Presidency in the reflexion carried out on ways and means on the possible evolution of ESARDA to address the new international and European non-proliferation and security challenges.

1st meeting of the ESARDA Reflexion Group RG2010
JRC Brussels, 21st January 2010

- Second), the internal meetings. The VTM WG meet at least twice a year, once during the annual meeting and once during the fall often in coordination with the C/S and IS working group which allows to have good exchanges on topics of common interest[5].
- Third) Several meetings or sessions have been organized to deals with a specific subject as:
  - Satellite imagery in collaboration with GMES (…) or LIMES (EUSC/Torrejon, October 2009) project members
  - Export Control in cooperation with JRC/ISPRA/IPSC and DG-TRADE and the US DOE (ISPRA, …2006 and 2008)
  - Environmental monitoring (Luxembourg, May 2008) in cooperation with IGSE[6] (*Regina Hagen*, iGSE Coordinator and *Martin Kalinowski,* Chair)
- Fourth), in cooperation with the International Safeguards Division of the INMM[7] and the local correspondent, the VTM working organizes the joint INMM symposium meeting ~120 experts to topical deals with legal and technical non-proliferation and security issues :
  - In 2003 at Como (It) with the JRC
  - In 2005 at Santa Fe (US) with the US Safeguards Support Programme
  - In 2008 at Tokyo (Jap) with the Japanese and Korean INMM chapters
  - In 2011 in France

- Fifth) active contribution to the INMM Annual Conferences

29Th ESARDA Annual Meeting
Aix en Provence, 2007

IS, C/S and IPSC members relaxe after the VTM meeting,
ISPRA, 10 November 2009

VI. Prospects and objectives of the VTM for 2010 and beyond

The prospect for the year 2010 of the VTM working group and members will be regarding the IAEA to support the Safeguards objectives of early detection of non compliance, the implementation of the Information driven safeguards, regarding the EU external security policy to support the EU strategy against WMD proliferation through instruments as the Instrument for Stability, the CBRN task force the GMES, to support as far as possible the definition & implementation of non-proliferation and disarmament treaties in particular the 2010 Non Proliferation Treaty Review Conference, inception of the negotiation of a Cut-off treaty, the preparation of the Entry into Force of the CTBT and the preparation of the On Site Inspection. As for the environmental treaty a post Kyoto reflexion will be conducted

Regarding domestic ESARDA activities, the VTM as a transverse working group will work to develop coordination with other ESARDA WG through the organisation of sessions of common interests and definition of joint projects and with EU Think tanks (Vertic, IPFM, Insap, ...) and INMM.

An important internal objective will be also to update the ESARDA web site VTM WG page as a tool of communication between members and archives of the past events. But the objective of utmost importance is to actively contribute to the work of the 2010 reflexion group on the evolution of ESARDA to give the association and its management the key of its future. The issue of the management of the membership has also to be tackle in line of the future outcomes of the ESARDA RG2010. How to mobilize the members on a common project and maintain the links between them as the origin of the members of the VTM is rather wide coming from a large span of laboratories, Academics and Institutions either National, European, Non European or individual and the issues addressed are also rather wide as described in section III. Regarding the participation of the VTM to seminars and conferences, the programme is the ESARDA Luxembourg symposium, the INMM annual conference and on the top of the agenda, the Quadrennial November IAEA Symposium on International Safeguards: Preparing for Future Verification Challenges[8]

VII. Organisation/management

The VTM is organized over a Management Team: Michel Richard (current Chair), Dr. Gotthard Stein (current vice-chair and former Chair), Louis-Victor Bril (Secretary), and.Pr. Rudolf Avenhaus. The management team will change mid 2012. Due to the wide scopes of issues addressed by the VTM and the range of its various activities, the members of the group come from a large span of origin in Europe and abroad (United States, Japan, Canada, Australia, Russia,…): laboratories & research centres, universities, industry, international organisations (as the IAEA), European institutions national regulation authorities and administrations, Think-tanks and Non Governmental Organisation. The complexity of the VTM issues made it necessary to create subgroups:

- On Environmental Monitoring (Chair: Dr. Martin Kalinowski).
- On Satellite Imagery (Chair: Dr. Bhupendra Jasani)
- On export control (Chair: Dr. Filippo Sevini)

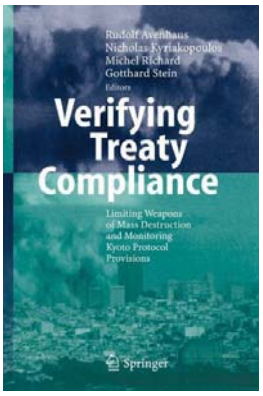
- From the work of the VTM on the proposal of Dr Whichello the Executive Committee of ESARDA has decided to create a new transverse working group dedicated to Novel Technologies and Approaches for IAEA Safeguards (NA/NT WG). This new group will work on technologies for the detection of undeclared activities with the support of the Novel Technology Unit of the Safeguards Department (Chair Harri Toiuvonen)

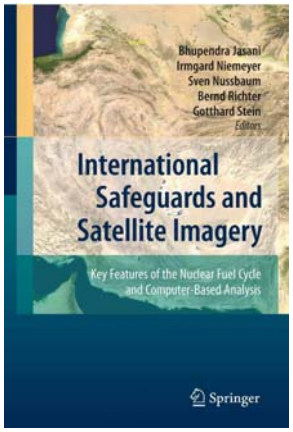
The VTM WG keeps close links with other governmental and non governmental institutions, in particular with INMM. Individual working group members volunteer to prepare discussion and working papers, subgroups have been established, conferences and meetings with special topics are performed. An important goal is to publish the results of major activities.

VIII. Publication & articles

The VTM WG has issued two publications:

- *Verifying Treaty Compliance: Limiting Weapons of Mass Destruction and Monitoring Kyoto Protocol Provisions, by (eds.) Rudolf Avenhaus, Nicholas Kyriakopoulos, Michel Richard and Gotthard Stein, Springer, July 2006.*





- *International Safeguards and Satellite Imagery: Key Features of the Nuclear Fuel Cycle and Computer-based Analysis, by (eds.) Bhupendra Jasani, Irmgard Niemeyer, Sven Nussbaum, Bernd Richter and Gotthard Stein, Springer, August 2008.*

A further book publication is planned for the complex field of proliferation resistance.

The VTM WG members make presentation at the INMM and ESARDA Symposium and publish articles in the Journal of Nuclear Material Management in the ESARDA Bulletin and other scientific publications.

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[2] 40 YEARS OF ESARDA: SAFEGUARDS MADE TO ORDER, Mrs. Elina Martikka, President of ESARDA, STUK - Radiation and Nuclear Safety Authority, Finland.

[3] Seminar on Modern Verification regimes: Similarities, Synergies and Challenges. Proceedings of the ESARDA annual seminar, Helsinki, Finland, 12-14 may 1998

[4] REPORT OF THE ESARDA REFLECTION GROUP 2000, M. Cuypers, JRC, Ispra On behalf of the Reflection Group. ESARDA BULLETIN, NO. 31.

[5] Presentations made during the VTM internal or specific meetings could be found in the restricted access section of the ESARDA web site either at: http://esarda2.jrc.it/about/organisation/working_groups.html, or at https://circa.europa.eu/Members/irc/securejrc/jrc_esarda/home.

[6] iGS: independent Group of Scientific Experts on the detection of clandestine nuclear weapons-usable materials production.

[7] INMM: International Nuclear Material Management.

[8] Vienna, Austria, 1–5 November 2010. Organized by the International Atomic Energy Agency In cooperation with the European Safeguards Research and Development Association (ESARDA) and the Institute of Nuclear Materials Management (INMM).

National R&D System in Hungary for Safeguards Related Research at Technical Support Organizations

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Abstract:

The main goal of the Hungarian safeguards related R&D activities is the support of international and national safeguards verification activities by developing equipment and technology for national safeguards verification purposes. A significant part of the R&D activities is covered by the Hungarian Safeguards Support Programme (HUN SP) to the International Atomic Energy Agency (IAEA), which was started 19 years ago. The goal of the HUN SP is to utilize our existing capacity in this field as much as possible and to contribute directly or indirectly to the strengthening the IAEA's safeguards regime in an organized way. Main R&D activities cover NDA methods for verification of nuclear material accountancy, detection of illicit trafficking of nuclear materials and development of new data acquisition methods. Moreover, DA methods have been also developed for environmental sampling of trace amounts of uranium and transuranium elements, and detailed investigation of seized nuclear materials.

Keywords: R&D, Technical Support Organisation (TSO), safeguards

1. Introduction

Generally the R&D activities are carried out by Technical Support Organizations (TSO) under the coordination of the Hungarian Atomic Energy Authority (HAEA). HAEA has been focused not only on balancing the resources between the TSOs but also to concentrate it to research fields of high national and international importance. The R&D activities are focusing on non-destructive (DA) and destructive (DA) analytical methods supporting national safeguards activities and nuclear forensics investigations by developing equipments and procedures. In this paper these activities and some of the most important results will be overviewed. Since a significant part of the safeguards related R&D activities is covered by the Hungarian Safeguards Support Programme (HUN SP) to the International Atomic Energy Agency (IAEA), the results of the HUN SP will be presented as well.

2. Technology developments in the frame of HUN SP

The technology development activities are partially financed by HAEA through the budget dedicated to the R&D of the national safeguards verification system. Our method development activities are focusing both on DA and on NDA techniques. During the last two decades the following major activities were carried out:

- Modification of IAEA Computer Codes for MTR Measurements (1994-1996)
- CdTe Based Portable Spent Fuel Attribute Tester (PSFAT) for the Paks NPP (1996)
- Tomographic Method for Verification of Irradiated Fuel Assemblies (FIN,SWE, HUN) (1999-2003)
- Verification of WWER-440 Absorber Assemblies and Co-60 Source Holders at Paks NPP (1999)
- Verification method and equipment of Pu in Pu-Be Neutron sources by Neutron Assay (2004-2007) [1]

- Software development for the prototype of the Tomographic Spent Fuel Detector System (2004-2009)

Several selected important results are described in details

2.1. Multiplicity Spectrometer Prototype

Multiplicity Spectrometer Prototype has been developed at the Institute of Isotopes of the Hungarian Academy of Sciences to support neutron coincidence data analysis for safeguards. Neutron coincidence counting is a widely adopted nondestructive assay (NDA) technique used in nuclear safeguards to measure the mass of nuclear material in samples. Nowadays, most neutron-counting systems are based on the original-shift-register technology, like the (ordinary or multiplicity) Shift-Register Analyser. The analogue signal from the He-3 tubes is processed by an amplifier/single channel analyser (SCA) producing a train of TTL pulses that are fed into an electronic unit that performs the time- correlation analysis. The development of prototypes of neutron-counting systems with PC-based processing have started in collaboration with European Union Joint Research Centre (EC JRC), Institut de Radioprotection et Sureté Nucléaire and USA Department of Energy Los Alamos National Laboratory (DOE LANL) Joint testing campaigns have been performed in the JRC PERLA laboratory, using different equipment provided by the partners.

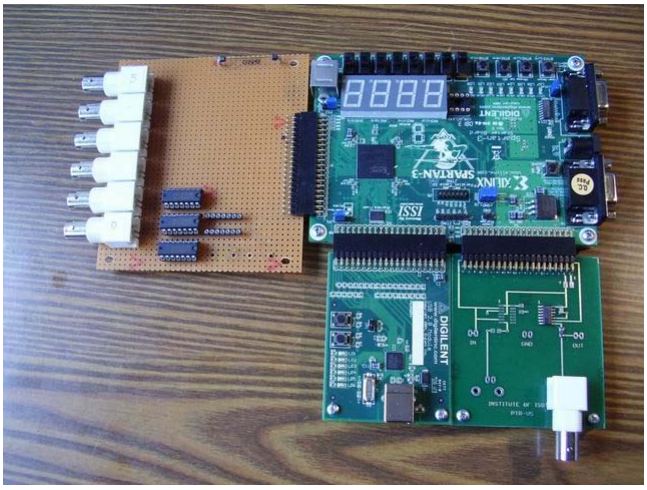


Figure 1. Novel multichannel reader for neutron coincidence measurements

One area of development is the use of high-speed PCs and pulse acquisition electronics that provide a time stamp (LIST-Mode Acquisition) for every digital pulse. The time stamp data can be processed directly during acquisition or saved on a hard disk. The latter method has the advantage that measurement data can be analysed with different values for parameters like predelay and gate width, without repeating the acquisition. Other useful diagnostic information, such as die-away time and dead time, can also be extracted from this stored data. A second area is the development of “virtual instruments.” These devices, in which the pulse-processing system can be embedded in the neutron counter itself and sends counting data to a PC, can give increased data-acquisition speeds. Either or both of these developments could give rise to the next generation of instrumentation for improved practical neutron-correlation measurements. [2]

2.2. Joining process to the IAEA Network of Analytical Laboratories (NWAL)

Hungary expressed its willingness to contribute to the work of the NWAL for analysis of environmental swipe samples collected by IAEA inspectors. During 2009-2010 the ICP-MS laboratory of the Institute of Isotopes of the Hungarian Academy of Sciences developed the required analytical methods, which fulfils the analytical performance criteria of IAEA (see Fig. 2.).

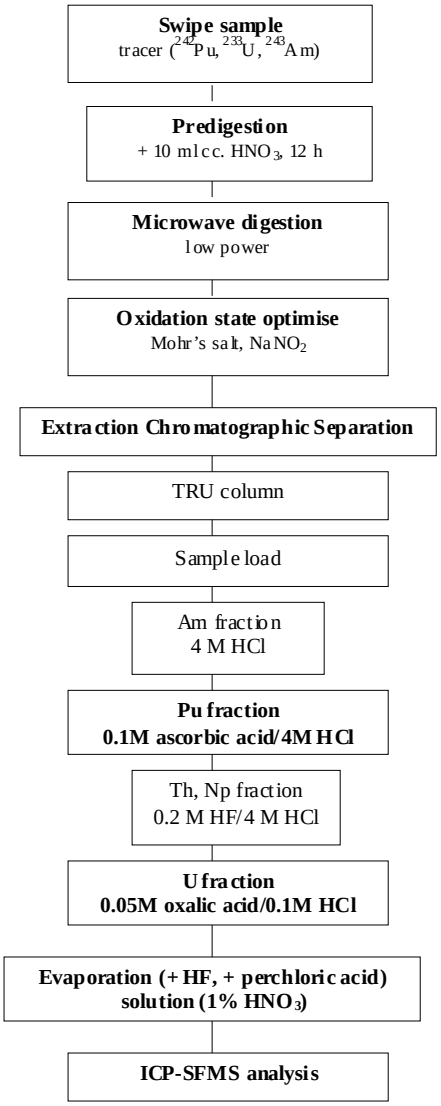


Figure 2. Schematic diagram of the sample preparation process

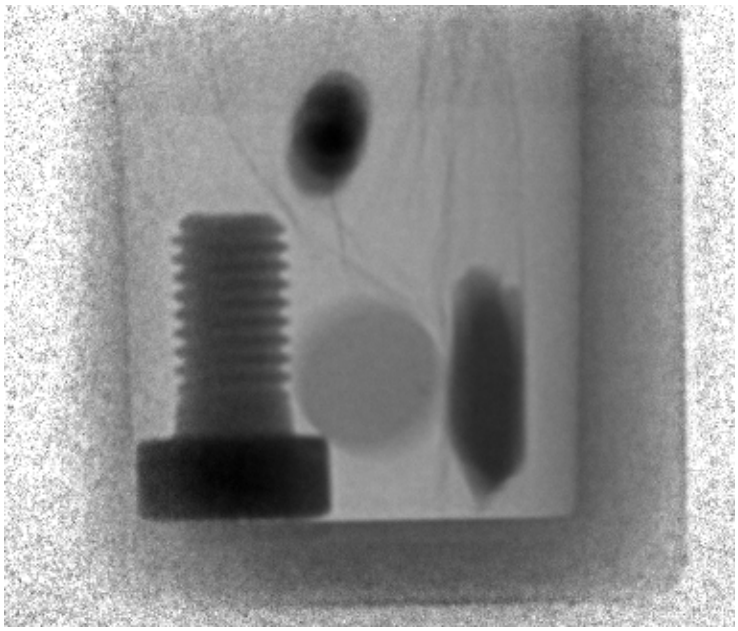
The method based on an extraction chromatographic separation and pre-concentration of the analytes of interest followed by ICP-MS analysis. Moreover, the Institute built up its own quality assurance system according to the requirements of the IAEA. Currently the analysis of the test samples sent by the IAEA is in progress.

3. Technology developments for national safeguards purposes

3.1. Prompt Gamma Activation Analysis for safeguards and nuclear forensics

Since the introduction of the Additional Protocol and the very much different new technology involved – like satellite imagery, information retrieval and processing, etc- a significant shift can be experienced in request for support from the traditional nuclear material verification methods towards new and novel technologies. Therefore Hungary has decided to take active part in development of these novel technologies. Application of Prompt Gamma Activation Analysis (PGAA) for the possible detection of massively sealed (hidden) nuclear materials is under development. Using this technique not only the nuclear materials can be detected inside of a "black-box", but also the mass and volume of the nuclear material can be estimated. Moreover, the analysis of other materials being present in the sample can

also be carried out (See Fig 3.). [3] This technique could be extremely useful for security and/or nuclear forensics purposes as well.



.**Figure 3.** Different type of materials inside a sealed container (uranium is present on the right-hand side

3.1. Laser ablation assisted ICP-MS methods

Another example for the new developments is the application of laser ablation inductively coupled plasma mass spectrometry (LA-ICPMS) for isotopic distribution determination and age-dating of nuclear materials based on $^{234}U/^{230}Th$ ratio. Laser ablation sample introduction offers several advantages e.g. the low sampling amount required for analysis (typically few µg) and the short measurement time. The aim of the investigation is the rapid and more comprehensive categorization of seized nuclear. On Figure 4 the surface of a nuclear based material can be seen after the ablation. [4-5]

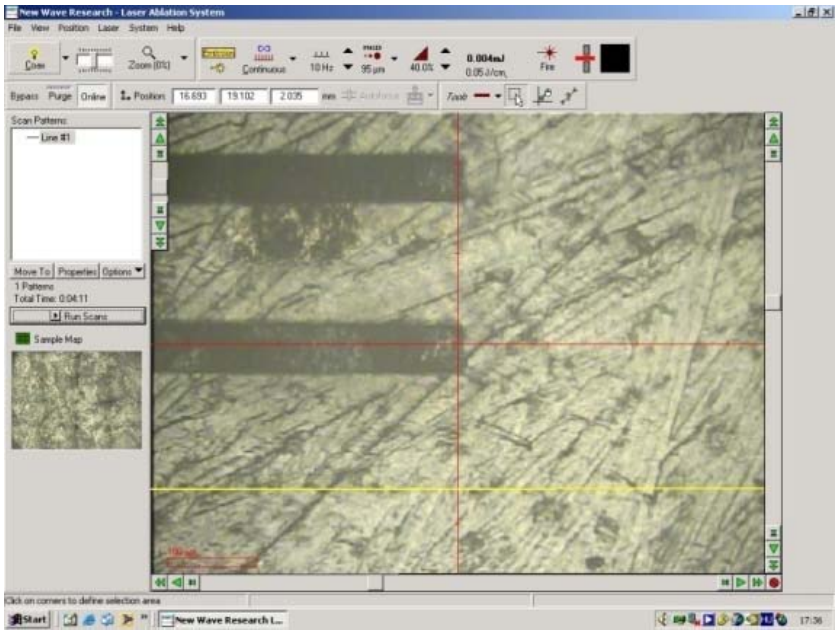


Figure 4. Uranium-oxide materials after LA-ICPMS measurements

4. Conclusion

Safeguards related R&D in Hungary is coordinated by HAEA and based on the methods and instruments developed by TSOs. A considerable part of R&D is covered by HUN SP to the IAEA, however several developments provided by TSOs are only for national safeguards purposes. Coordinating R&D activities HAEA pays attention to careful balancing of different development directions and always focuses on up-to-date knowledge and trends in safeguards related scientific fields. Additionally to traditional safeguards methods Hungary has been actively participating in the development and testing of novel safeguards technologies.

5. Acknowledgements

The Hungarian Atomic Energy Authority greatly acknowledges the valuable effort and work that are devoted by all the participants of the HUN SP and the TSOs, especially that of the Paks Nuclear Power Plant, the Institute of Isotopes and the KFKI Atomic Energy Research Institute of the Hungarian Academy of Sciences, the Institute of Nuclear Techniques of the Budapest University of Technology and Economics, the MECSEK-ÖKO Ltd, the Public Limited Company for Radioactive Waste Management and the Institute of Isotopes Ltd.

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An improved vulnerability assessment of safeguarding a geological repository

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Abstract

In a previous publication [1] the principles of a vulnerability assessment of the safeguards approach for a geological repository were elaborated and an illustrative example for optimising the safeguards approach for several diversion paths was given, based on the parameters detection probability, false alarm probability and delay time (between diversion and detection).
For an improved optimisation the cost of implementing the safeguards approach has been included as a parameter in the vulnerability assessment.
Furthermore the considered diversion paths and safeguards measures have been evaluated and updated according to the state-of-the-art of safeguards for a geological repository.

Keywords: vulnerability assessment, geological repository, safeguards

1. Introduction

In the framework of the ASTOR task, a joint Support Programme task to the IAEA with the purpose to discuss and develop the safeguards approach for geological repositories, the focus lies at present on the Finnish design of a geological repository since it is the first actually being built. The IAEA has presented a draft safeguards approach for a geological repository that defines the safeguards measures to be applied to such an installation. The draft safeguards approach is an integrated safeguards approach, taking into account the fact that the host country has signed the Additional Protocol and that the IAEA has drawn a broader conclusion with respect to the absence of undeclared activities in the host state.

Diversion scenarios for a geological repository have been developed in the past. Based on these diversion scenarios, the proposed safeguards measures can be evaluated with respect to their effectiveness and efficiency. In this paper we use an approach based on multi-attribute value theory for the evaluation and analysis of the proposed safeguards measures.

2. Description of the geological repository

The description of the reference geological repository was developed during SAGOR's phase 1 and is based on Wuschke [2]. The description is divided in two parts: a part describing the physical properties of the site and a part describing the activities at the site. In a previous paper [1] a summary was given. For the full description the reader may consult [2,3]. Present IAEA activities are focussing on the Finnish design of a geological repository, since this is actually being commissioned. In this chapter we will give a schematic overview of the Finnish design, which does not deviate from the SAGOR design to a large extent.

2.1. Physical properties of an open geological repository

The Finnish repository is excavated in granite. The Finnish repository can be accessed by vehicles via a ramp. Additionally there is a shaft for access for personnel and two ventilation shafts. Large arrays of disposal rooms will be excavated for the emplacement of the spent fuel canisters. These canisters contain 4 or 12 spent fuel assemblies, dependent on their origin. They consist of a cast iron insert to fix the fuel assemblies with a copper outside of 50 mm. About 2800 canisters are expected to be emplaced in the repository, either horizontally or vertically.

2.2. Extended description of the material flow to the repository

Figure 1 gives a schematic overview of the material flow concerning spent fuel assemblies from the reactor to the geological repository.

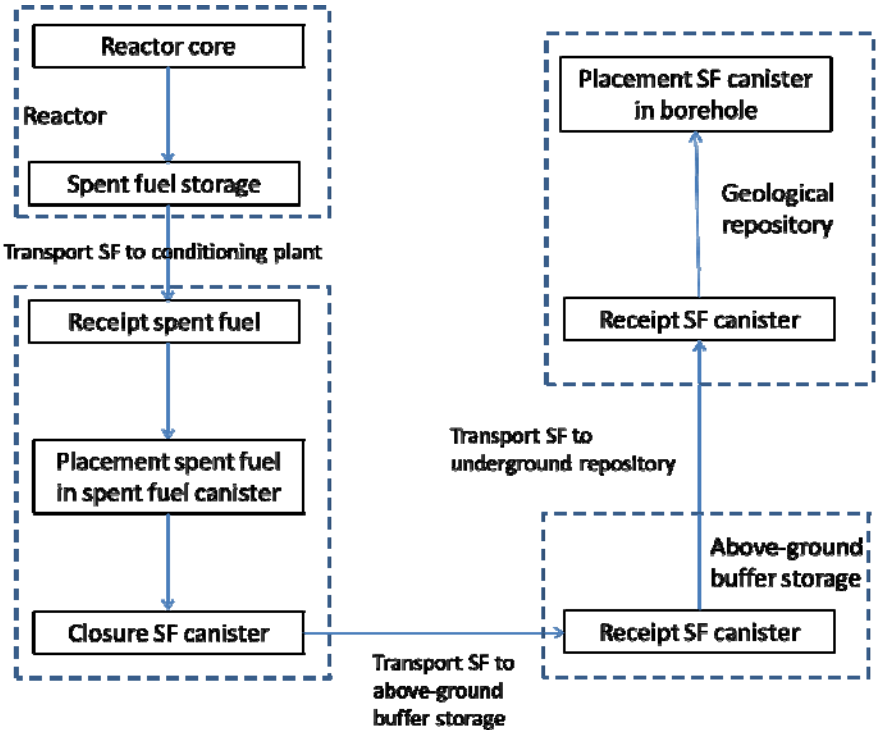


Figure 1: Overview of the material flow concerning spent fuel assemblies from the reactor to the geological repository

3. Diversion scenarios and associated anomalies

The declared nuclear material in a geological repository consists of spent fuel assemblies that are packed in a disposal canister. Diversion will consists of the diversion of a whole canister or part of its content in the form of a spent fuel assembly or one or more irradiated fuel pin(s).
Diversion will take place before the canister enters the geological repository or after it has entered the geological repository. For simplicity of the analysis no distinction is made where a diversion will take place while the canister is in the repository (buffer zone, tunnel or borehole). When the diversion takes place in the repository, the canister or parts of its content (fuel assembly, fuel pins) will be transported back to the surface via existing shafts/ramps or via unreported tunnels. Another possibility is the presence of a small reprocessing facility underground, so that only small quantities of Pu have to be transported to the surface.
In the case of both an open and a closed repository, canisters have to be retrieved via unreported tunneling activities.

Associated anomalies

Diversion of loaded canister before entrance repository
- Falsification of Nuclear Material Accountancy (NMA)
- Missing canister
- Presence dummy canister
- Environmental Sampling (ES) signatures for reprocessing activities

Diversion of canister after entrance repository
- Unreported transport of loaded canister to surface
    - Via entrance/exit shaft/ramp
    - Via other existing shafts
    - Via unreported shafts/ramps
- Radiation signatures of transport casks coming from repository
- Unreported tunneling activities
- ES signatures for reprocessing activities

Diversion of fuel assemblies or fuel pins after entrance repository
- Unreported transport of fuel assembly or fuel pins to surface
    - Via entrance/exit shaft/ramp
    - Via other existing shafts
    - Via unreported shafts/ramps
- Radiation signatures of transport casks coming from repository
- Unreported tunneling activities
- Presence of hot cell facilities in repository not according to Design Information (DI)
- Presence of rooms not according to DI
- ES signatures for reprocessing activities

Reprocessing in repository
- ES signatures for reprocessing activities
- Presence of rooms not according to DI
- Presence of reprocessing facility in repository (hot cells with specific reprocessing equipment)

Diversion of canisters via undeclared tunneling from open or closed repository
- Unreported tunneling activities
- ES signatures for reprocessing activities

4. Integrated safeguards approach for a geological repository

Based on a draft safeguards approach for a geological repository [4], a set of safeguards measures was defined for use in the analysis of this paper. The set safeguards measures consists of:

- NMA
- Containment and Surveillance (C/S), application of seals to spent fuel canisters
- C/S, camera surveillance on movements of spent fuel canisters
- Monitoring, based on e.g. radiation monitoring, canister identification, directional movement monitoring
- Design Information Verification (DIV), based on:
    - In situ verification
    - Satellite Imagery
    - Geophysical Monitoring

This set differs in a few aspects from the set used in our previous study [1]. In addition to this study we will evaluate all diversion scenarios for a geological repository.
Most combinations of the above-mentioned safeguards measures were evaluated. NMA was included in all combinations and the in-situ DIV was included in all DIV activities. In the following we discuss shortly the safeguards measures and their effectiveness in relation to the various diversion scenarios.

Nuclear Material Accountancy (NMA)

Nuclear Material Accountancy is the basis of safeguards. Without NMA an inspection cannot be performed since the inspector will have no reference to the amount of nuclear material that has to be inspected.

Containment and Surveillence (C/S) with Seals (SE)

The application of seals has the purpose to provide Continuity of Knowledge (CoK) about the nuclear material under seal. The effectiveness of the seal depends on the type of seal and the item that is under seal. The latter is in this case a spent fuel canister. Normally a closed and sealed spent fuel canister could not be opened without noticing tampering of the seal or observing unexpected welds on the canister. However, the Swedish SKB has shown an example of a weld that cannot be distinguished from unwelded canister material (in this case copper). This provides a diversion scenario that cannot be detected by the application of seals.

Containment and Surveillence (C/S) with optical Camera surveillance (CA)

Camera surveillance or optical surveillance has also the purpose of providing Continuity of Knowledge of the nuclear material under surveillance. Again the items under surveillance are spent fuel canisters. Camera or optical surveillance can be applied to a static situation or a situation where there are movements of canisters. The latter is the most likely, since long-term storage of spent fuel canisters is not foreseen until the canisters are finally deposited in a borehole. This means that multiple areas have to be covered by optical surveillance in order to conserve the CoK of the canisters. This implies the use of multiple cameras and the condition that none of these cameras fails during the observation period.

Dual C/S

Dual C/S implies the combination of two independent C/S techniques, in this case the use of seals and optical surveillance. It is more robust than the use of single C/S methods separately.

Monitoring (MON)

Monitoring has the same purpose of C/S, i.e. providing CoK of the monitored nuclear material (the spent fuel canisters). Monitoring will provide at most the same assurance that no nuclear material is diverted as optical surveillance does for the above-ground case. For the entrances of the geological repository monitoring (especially radiation monitoring) is considered more robust since it can detect nuclear material coming out of the repository. The level of assurance depends on the type of monitoring and the specific design of the repository.

Design Information Verification (DIV)

Whereas the detection of an above-ground diversion of nuclear material from spent fuel canisters is mainly dealt with by application of safeguards measures like C/S or monitoring, the diversion and possible processing (hot cells for dismantling and reprocessing of spent fuel) of canisters or undeclared tunnelling activities should be detected by application of DIV. Several types of DIV are envisaged:

In-situ verification (DIVV) will verify the absence of reprocessing activities or undeclared tunnelling. Depending on how well the diverter is able to hide these activities, in-situ verification may well or not detect them.
Satellite Monitoring (DIVSI) will verify undeclared tunnelling activities in the neighbourhood of the site of the geological repository. In view of the fact that the present tunnelling activities at Olkiluoto for the Finnish repository do not show very clear evidence on the satellite images, one may conclude that satellite imagery is not a fully robust method to detect these activities.
Geophysical Monitoring (DIVGM) will also verify undeclared tunnelling activities. Tests have shown that this method, applied unattendedly for passive monitoring, will be able to detect undeclared tunnelling activities.

5. An approach to vulnerability analysis of a geological repository

5.1 General framework

The vulnerability of a protected system with respect to an intrusion could be seen as a function of the susceptibility to the intrusion scenario, the resilience (capacity of the sytem to function when some of the protection components fail) and the environment. One can notice in the literature that the concept of vulnerability is multidimensional, most often the indicators used being qualitative.

Based on INFCIRC/153, the technical purpose of safeguards is: "…*the timely detection of diversion of significant quantities of nuclear material from peaceful nuclear activities to the manufacture of nuclear weapons or of other nuclear explosive devices or for purposes unknown, and deterrence of such diversion by the risk of early detection*."

For safeguards aims, the likelihood of a scenario is not of particular relevance since all diversion scenarios should be covered equally well by the safeguards approach, regardless of e.g. budgetary implications. A diverter will probably choose for the scenario with the lowest detection probability if this would exist.

The vulnerability of a geological repository to a given diversion scenario may be expressed drawing on the concept of vulnerability as employed for characterisation of critical infrastructures. In Ezell [5], the vulnerability of a system to unauthorized actions (inclusing terrorism) can be defined as a multi-dimensional concept depending on: deterrance (measures implemented that are perceived as difficult to defeat), probability of detection, delay time (time while an element of a sytem can delay entrance or exit from the protected area) and reponse (time to repond to a threat).

In the context of safeguards, the following indicators are proposed:

C_1: Detection robustness
In the previous paper detection probability was used as an indicator. This concept was considered not sufficient since it does not take into account the robustness of a safeguards measure against willful falsification. Detection probability is considered as based on random events like technical failures. Since the consequences of these random events are reduced as much as possible by the equipment design, the detection probability will often approximate 100% and is therefore not very useful as an indicator.
We introduce the concept detection robustness as a combination of detection probability and robustness to falsification. The value that is allocated means that even in case of falsification, this is the probability that a diversion will be detected by the safeguards system.

C_2: False alarm probability
The probability that a safeguards measure will trigger an alarm without a real diversion taking place. It is the purpose of the inspector to keep the false alarm probability as low as possible.

C_3: Delay time
The delay time is the time between a diversion and its discovery. It is the aim of the safeguards system to keep the delay time as low as possible ("risk of early detection").

The vulnerability V of the repository in the context of a safeguards strategy *S* and the diversion scenario *d* is defined as:

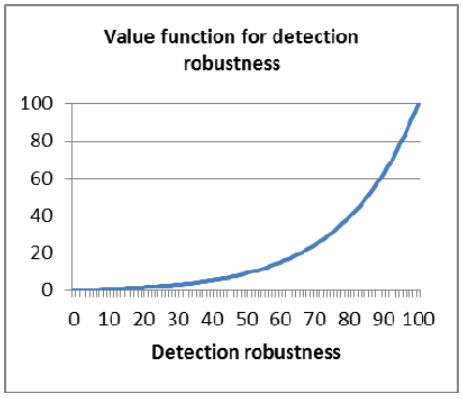
$$V_d(S) = 100 - \sum_{i=1}^{3} w_i \cdot f_i(C_i(S,d)), \quad \text{with } \sum_{i=1}^{3} w_i = 1.$$

The scale used to evaluate vulnerability is 0-100, with a score 0 corresponding to a system that is not vulnerable and a score of 100 corrresponding to a system that is completely vulnerable.

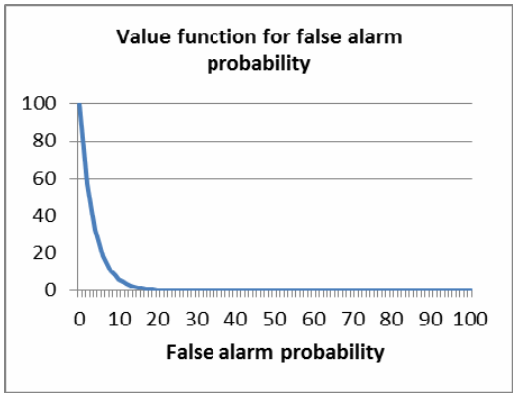
The weights w_i are related to the importance of the different vulnerability indicators. They have been derived based on pairwise comparisons of the three indicators, with the help of the Web-HIPRE software [6], via the so-called AHP method (Analytical Hierarchical Process) by Saaty [7]. Detection of a diversion is the most important since without the detection of a diversion the main safeguards goal is

not achieved. The delay time has been considered more important than the false alarm probability. The delay time is directly connected to "timely detection", whereas a higher false alarm probability will cause some reverification activities and has therefore mainly budgettary implications, assuming reverification activities can be performed appropriately. The derived weights were 0.714 for the detection probability, 0.223 for the delay time and 0.063 for the false alarm probability.

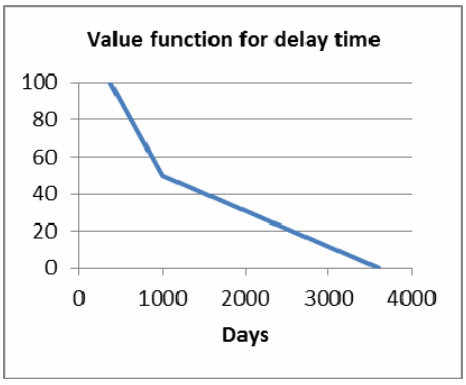
The function f_i is a value function associated to indicator C_i, and expresses the degree of fullfillment of the safeguards objective by the particular value $C_i(S,d)$ atained for safeguards strategy S and diversion scenario d. This function is in fact a representation of the proliferation resistance of the system (the opposite of vulnerability). For instance, a delay time of less than 365 days can be considered fully satisfactory (f_i will have a value of 100), whereas a delay time of more than 10 years is completely unsatisfactory (f_i will have a value 0), with a linear, piecewise linear or exponential dependency in between these extreme values. Fig. 3a,b,c presents the value functions corresponding to the three vulnerability indicators.



2a: exponential function



2b: exponential function



2c: piecewise linear function

Figure 2: Value functions for the three vulnerability indicators

Similar to [1], for the detection robustness, we penalised scores that are below 50%; these were considered as having a low value (less than 10 on a scale from 0 to 100). Only for detection probabilities higher than 85% the value function corresponding to the detection robustness becomes higher than 50%.

For the false alarm probability we assumed that medium and high false alarm rates, i.e. of more than 50%, are not tolerable for safeguards inspections (i.e. value ~0). These false alarm rates would undermine the confidence in the safeguards inspections and would increase the costs too much due to the necessity to repeat often (parts of) inspections.

For the delay time we assumed that 1 year is perfectly acceptable (value 100). A delay time of 3 years (~1000 days) may still be considered acceptable (value 50 on a scale from 0 to 100), whereas a delay time of more than 10 years is certainly not acceptable for safeguards.

5.2 Evaluation of the safeguards measures

In the annex of this paper we provide table A1 with the data that were used for the evaluation of the safeguards measures. We provide only the data for the first scenario, since providing all the data falls beyond the scope of this paper.

NMA alone is not considered effective to detect diversion since the accountancy can be changed without being noticed if the nuclear material is not verified at all. Therefore, the detection robustness is given a value of 0%.

Seals and cameras applied individually have been considered to provide 50% detection probability for a diversion. As they are considered as independent methods, their combined detection robustness is 75%.

Monitoring (other than optical) has been considered as less effective than optical monitoring since only a partial aspect of the monitored item is considered. Monitoring in combination with seals is considered as the use of two independent methods, while monitoring in combination with cameras is considered as the use of partly dependent methods. This results in a slightly smaller value for the detection robustness for the latter.

For each set of measures we have evaluated apart from the vulnerability indicators also the associated costs:

C_4: Costs
Costs have been expressed in person days of inspection (pdi). They are based on the time spent by the inspector to gather and analyse the data and perform simple maintenance. This means that investment costs have not been taken into account.

The results of the assessment for the five diversion scenarios are presented in Table A2 of the annex. For each diversion scenario we highlight the best achievable performance (lowest vulnerability score). For each set of measures the highest vulnerability score and the maximal distance from the best achievable score are given.

6. Discussion

As in our previous paper, we would like to remark that the scores given to the various vulnerability indicators do not necessarily represent the whole safeguards truth. Our purpose here is merely to show the methodology and to provide an example of a vulnerability assessment for use by the IAEA.

From the results shown in Table A2 (last row) we can see that the lowest scores of the vulnerability for the five considered diversion paths are 49%, 49%, 66%, 16% and 15%, resp. We see that the third diversion path has the highest vulnerability: 66%. This is due to the fact that none of the applied measures is considered to detect in a highly efficient way a diversion of a fuel element or a few fuel pins from the repository to an above-ground facility.

From this observation it can be concluded that safeguards measures should be developed or modified in order to detect with higher accuracy a diversion of few fuel pins from the repository. This can be done by improving the detection limit of the radiation monitoring equipment that is installed at the entrances of the repository. An alternative conclusion, in view of the first remark in this section, could be to reconsider our analysis and to see whether the given scores for the detection robustness are valid or need to be adjusted.

Considering the highest score for the vulnerability being 66%, we then can derive from the 7^{th} column of the results in Table A2 the optimal cost for the inspection. This turns out to be 5 pdi for the combination of NMA, DIVV, CA and MON. Compared to the highest cost value of 10 pdi for a vulnerability of 66%, this results in a reduction of 50% of the inspection effort. A drawback for this optimisation is that the vulnerability for three diversion paths is 66%, while for 10 pdi the vulnerability for only one diversion path is 66%.

From the same column we can see that accepting a slightly higher vulnerability of 5% (from 66% to 69%) will result in a further reduction of inspection effort of 20% (from 5 to 4 pdi).

Apart from looking at the highest vulnerability for each safeguards strategy ("worst case scenario") it is informative to look at the deviation from the best achievable score. This provides a more detailed information than looking only at the worst case scenario and may rule out, for instance, a combination such as NMA, DIVV, MON, CA (#15) since it deviates quite strongly from the best value achievable. The latter reasoning highlights instead strategies such as #32 (NMA, DIVV, DIVGM, MON, SE, CA) which is nearly best in all diversion scenarios, but has a 20% lower cost than #40 (NMA, DIVV, DIVSI, DIVGM, MON, SE, CA) which is optimal with respect to vulnerability for all scenarios.

7. Conclusions

Vulnerability assessment of the application of various sets of safeguards measures to various diversion scenarios provides an insight in the most vulnerable diversion path and shows where improvements can be made efficiently to reduce vulnerability.

The assessment can furthermore show what combination of safeguards measures provides the most cost-effective set of measures.

In this example a combination of Nuclear Material Accountancy, In-Situ Design Information Verification, Camera Surveillance and Radiation Monitoring provides the most cost-effective way to obtain the lowest possible vulnerability. The results obtained are strongly dependent on the given scores for the vulnerability indicators. It is strongly advised to review these by experienced IAEA inspectors.

The example showed that a diversion of a fuel element or fuel pins from the repository provides the highest vulnerability for the safeguards system.

Further work will focus on a sensitivity analysis to provide insight in the uncertainties of the assessment methodology.

8. Acknowledgements

Support for this work from NIRAS/ONDRAF, the Belgian National Institute for the Management of RadioActive Waste and Fissile Materials, is highly appreciated.

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Annex. Data used for the evaluation of safeguards measures

Measures	Detection robust-ness (%)	False alarm prob. (%)	Delay time (days)	Cost (pdi)	Measures	Detection robust-ness (%)	False alarm prob. (%)	Delay time (days)	Cost (pdi)
NMA	0	0	3600	0.5	NMA, DIVV, DIVSI, MON	40	5	30	5
NMA, SE	50	1	360	1	NMA, DIVV, DIVSI, MON, SE	70	6	195	5.5
NMA, CA	50	5	30	2.5	NMA, DIVV, DIVSI, MON, CA	60	10	30	7
NMA, SE, CA	75	6	195	3	NMA, DIVV, DIVSI, MON, SE, CA	80	10	30	7.5
NMA, MON	40	5	30	2.5	NMA, DIVV, DIVGM	0	0	3600	3
NMA, MON, SE	70	6	195	3	NMA, DIVV, DIVGM, SE	50	1	360	3.5
NMA, MON, CA	60	10	30	4.5	NMA, DIVV, DIVGM, CA	50	5	30	5
NMA, MON, SE, CA	80	10	30	5	NMA, DIVV, DIVGM, SE, CA	75	6	195	5.5
NMA, DIVV	0	0	3600	1	NMA, DIVV, DIVGM, MON	40	5	30	5
NMA, DIVV, SE	50	1	360	1.5	NMA, DIVV, DIVGM, MON, SE	70	6	195	5.5
NMA, DIVV, CA	50	5	30	3	NMA, DIVV, DIVGM, MON, CA	60	10	30	7
NMA, DIVV, SE, CA	75	6	195	3.5	NMA, DIVV, DIVGM, MON, SE, CA	80	10	30	7.5
NMA, DIVV, MON	40	5	30	3	NMA, DIVV, DIVSI, DIVGM	0	0	3600	5
NMA, DIVV, MON, SE	70	6	195	3.5	NMA, DIVV, DIVSI, DIVGM, SE	50	1	360	5.5
NMA, DIVV, MON, CA	60	10	30	5	NMA, DIVV, DIVSI, DIVGM, CA	50	5	30	7
NMA, DIVV, MON, SE, CA	80	10	30	5.5	NMA, DIVV, DIVSI, DIVGM, SE, CA	75	6	195	7.5
NMA, DIVV, DIVSI	0	0	3600	3	NMA, DIVV, DIVSI, DIVGM, MON	40	5	30	7
NMA, DIVV, DIVSI, SE	50	1	360	3.5	NMA, DIVV, DIVSI, DIVGM, MON, SE	70	6	195	7.5
NMA, DIVV, DIVSI, CA	50	5	30	5	NMA, DIVV, DIVSI, DIVGM, MON, CA	60	10	30	9
NMA, DIVV, DIVSI, SE, CA	75	6	195	5.5	NMA, DIVV, DIVSI, DIVGM, MON, SE, CA	80	10	30	9.5

Table A1: Input data for diversion path 1: transport to reception area

	Measures	V1	V2	V3	V4	V5	Vworst	Cost	Max dist from opt vuln
1	NMA	94	94	94	94	94	94	1	79
2	NMA, SE	66	94	94	94	94	94	1	79
3	NMA, CA	69	69	73	94	94	94	3	79
4	NMA, SE, CA	54	69	73	94	94	94	3	79
5	NMA, MON	72	58	69	94	94	94	3	79
6	NMA, MON, SE	59	58	69	94	94	94	3	79
7	NMA, MON, CA	66	49	66	94	94	94	5	79
8	NMA, MON, SE, CA	49	49	66	94	94	94	5	79
9	NMA, DIVV	94	94	94	16	66	94	1	51
10	NMA, DIVV, SE	66	94	94	16	66	94	2	51
11	NMA, DIVV, CA	69	69	73	16	66	73	3	51
12	NMA, DIVV, SE, CA	54	69	73	16	66	73	4	51
13	NMA, DIVV, MON	72	58	69	16	66	72	3	51
14	NMA, DIVV, MON, SE	59	58	69	16	66	69	4	51
15	NMA, DIVV, MON, CA	66	49	66	16	66	66	5	51
16	NMA, DIVV, MON, SE, CA	49	49	66	16	66	66	6	51
17	NMA, DIVV, DIVSI	94	94	94	16	65	94	3	50
18	NMA, DIVV, DIVSI, SE	66	94	94	16	65	94	4	50
19	NMA, DIVV, DIVSI, CA	69	69	73	16	65	73	5	50
20	NMA, DIVV, DIVSI, SE, CA	54	69	73	16	65	73	6	50
21	NMA, DIVV, DIVSI, MON	72	58	69	16	65	72	5	50
22	NMA, DIVV, DIVSI, MON, SE	59	58	69	16	65	69	6	50
23	NMA, DIVV, DIVSI, MON, CA	66	49	66	16	65	66	7	50
24	NMA, DIVV, DIVSI, MON, SE, CA	49	49	66	16	65	66	8	50
25	NMA, DIVV, DIVGM	94	94	94	16	19	94	3	45
26	NMA, DIVV, DIVGM, SE	66	94	94	16	19	94	4	45
27	NMA, DIVV, DIVGM, CA	69	69	73	16	19	73	5	20
28	NMA, DIVV, DIVGM, SE, CA	54	69	73	16	19	73	6	20
29	NMA, DIVV, DIVGM, MON	72	58	69	16	19	72	5	23
30	NMA, DIVV, DIVGM, MON, SE	59	58	69	16	19	69	6	10
31	NMA, DIVV, DIVGM, MON, CA	66	49	66	16	19	66	7	17
32	NMA, DIVV, DIVGM, MON, SE, CA	49	49	66	16	19	66	8	4
33	NMA, DIVV, DIVSI, DIVGM	94	94	94	16	15	94	5	45
34	NMA, DIVV, DIVSI, DIVGM, SE	66	94	94	16	15	94	6	45
35	NMA, DIVV, DIVSI, DIVGM, CA	69	69	73	16	15	73	7	20
36	NMA, DIVV, DIVSI, DIVGM, SE, CA	54	69	73	16	15	73	8	20
37	NMA, DIVV, DIVSI, DIVGM, MON	72	58	69	16	15	72	7	23
38	NMA, DIVV, DIVSI, DIVGM, MON, SE	59	58	69	16	15	69	8	10
39	NMA, DIVV, DIVSI, DIVGM, MON, CA	66	49	66	16	15	66	9	17
40	NMA, DIVV, DIVSI, DIVGM, MON, SE, CA	49	49	66	16	15	66	10	0
	Best achievable score	**49**	**49**	**66**	**16**	**15**	**66**	**1**	**0**

Table A2: Resumative table: vulnerability scores for the five diversion scenarios and costs of safeguards measures

A green colouring indicates scores close to best achievable score, whereas a red colouring indicates less satisfactory strategies.

Nuclear material importation and exportation notification process in France

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Abstract:

International safeguards agreements frequently impose an advance notification of nuclear material transfers to be sent by the operators and/or by the national Authorities. With the evolution of nuclear partnerships and the entry into force of the 302/2005 EURATOM regulation, the number of notifications declared by the French operators has considerably increased. Moreover, in France, three different legal scales must be respected: national bilateral agreements, like France/Australia or France/Japan, EAEC agreements, like EURATOM/Canada or EURATOM/USA, and the IAEA agreements, like INFCIRC/290 and INFCIRC/207 for France. In order to help the French operators to comply with their legal obligations, IRSN/DEND/SACI have designed a website which enables them to write and transmit automatically the notifications to the French Authorities, including all the required information. In parallel, an exhaustive notification guide explains to the implied persons the safeguards organization, the technical specificities of each agreement into force in France and in the European Union, the criteria to determine which regulation applies to a transfer, and the technical ways to send the notifications to the national Authority, which will transfer them to the relevant interlocutor (EURATOM, IAEA or Ministry of Foreign Affairs).

This article aims at raising up a panorama of the international agreements dispositions linked to the advance notification system, explaining the national organization to ensure that the international commitment of France are respected - and the key points of such process - and presenting the technical means given to the operators, like the unified notification form or the website PIMENT, to help them in writing their nuclear material transfer notifications.

Keywords: Agreement; Notification; Nuclear material; Safeguards; Transfer

1. Introduction

The efficiency of the safeguards global system relies on declaration of the operations on nuclear material, ascertained by an independent verification regime. As a consequence, the nuclear material transfers verification is a key activity for the efficiency of safeguards. It is clearly a complementary and indispensable element to improve the confidence in the operators NMAC system, the SSAC and RSAC.

France, which has always promoted the implementation of the non-proliferation regimes and the development of the peaceful uses of atomic energy, concluded with the IAEA and the EAEC a voluntary safeguard agreement completed by an additional protocol and the adhesion to the Agency agreements on sensible material transfers, like INFCIRC/207 or INFCIRC/415. The EURATOM regulation requires too the advance notification of imports and exports. In parallel, France has concluded many bilateral cooperation agreements with third States, which may require information on nuclear material transfers.

For reminding, the advance notification goal is to:

- Enable the verification (after reception or before shipment) of nuclear materials by the European Commission and the IAEA.
- Track nuclear materials transfers “on real time” (contrary to the accountancy system).
- Obtain prior consent from third States if it is required in cooperation agreements.

This article deals with the advance notification process in France, its organization and specificities, and the means to fully comply with these agreements and regulations.

2. International agreements commitments and structure

Any transfer of nuclear material from one country to another (in the European Union or not) must respect the provisions of several agreements. Four legal categories should be highlighted:

- Bilateral agreements concluded by France.
- The 302/2005 regulation [2].
- EURATOM agreements (concluded under article 101 of the EURATOM Treaty [1]), with the subsequent provisions and administrative arrangements.
- IAEA agreements (INFCIRC/207 [3] and INFCIRC/290 [4]).

In the European Union, the legal framework regarding nuclear materials is more comprehensive than the IAEA framework described in INFCIRC/153:

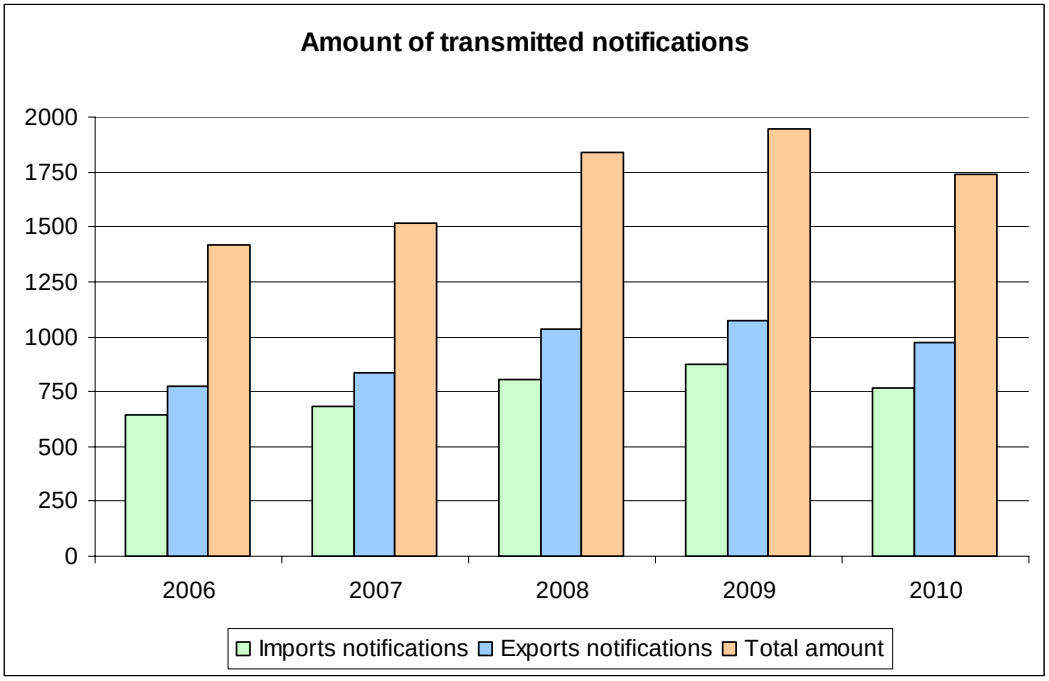
- Article 197 of the EURATOM Treaty [1]:
    - Special fissile materials (“plutonium 239; uranium 233; uranium enriched in uranium 235 or uranium 233; and any substance containing one or more of the foregoing isotopes and such other fissile materials as may be specified by the Council”).
    - Uranium enriched in uranium 235 or uranium 233.
    - Source materials.
    - Ores.
- EURATOM / USA [5] and EURATOM / Canada [6]: Deuterium

It is evident that any element, even if it is not a fissile, fertile or fusible material, can be subject to a bilateral agreement if accepted by the signatories. In the same time, these agreements contain frequently different or incompatible provisions. Figure 1 gives an example of difference between 302/2005 [2], INFCIRC/290 [4], and EURATOM / Australia [6] for exports.

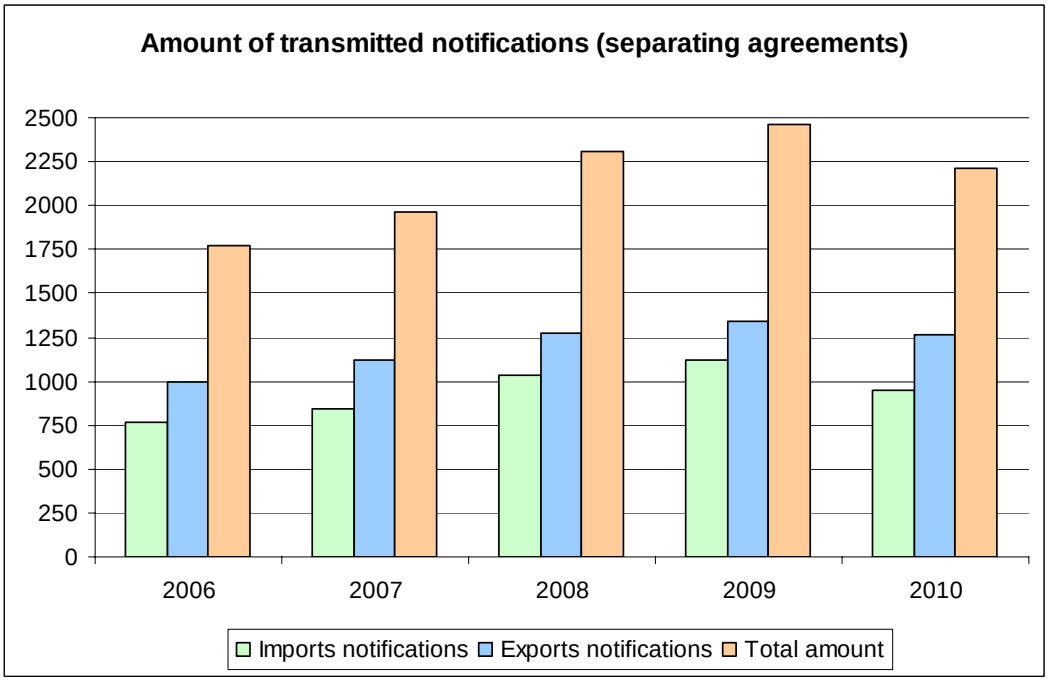
Provision	302/2005	INFCIRC/290	EURATOM / Australia
Delay for advance notification	10 working days before packing	10 working days before shipment	Depends on recipient country (10 days before packing or more for unlisted countries)
Confirmation of export	No	No	Yes (within 4 days)
Notification at 1st gram of material transferred	No	No	Yes
Effective kg for shipment	Yes	Yes	Yes
Effective kilogram in any consecutive period of twelve months	Yes	No	Yes
Recipient country	All	All	9 listed countries with generic prior consent
Use limitation	No	No	Depends on recipient country

Table 1: Differences in provisions for 302/2005 [2], INFCIRC/290 [4], and EURATOM / Australia [6].

Due to these numerous cooperation agreements and regulations, and since the entry into force of the 302/2005 regulation [2] which imposes a notification of all transfers inside / outside France (about +75% of notifications - previously, advance notification where required only for exports inside / outside the EU), the amount of notifications has considerably increased.



Graph 1: Amount of transmitted notifications since 2006.



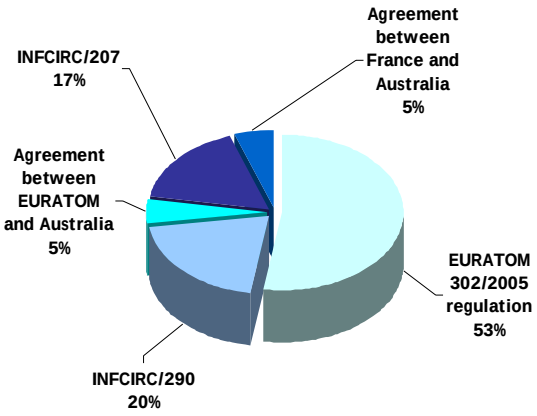
Graph 2: Amount of transmitted notifications by separating agreements since 2006.

The second graph shows the amount of notifications by agreement: the accountancy method is to count one notification for each agreement. For example, the notification of a transfer for Australia is

counted as two advance notifications, one for the 302/2005 regulation [2] and one for the EURATOM/Australia agreement [10]. The evolution is similar to the previous graph, and the number of notifications in percentage is quite stable.

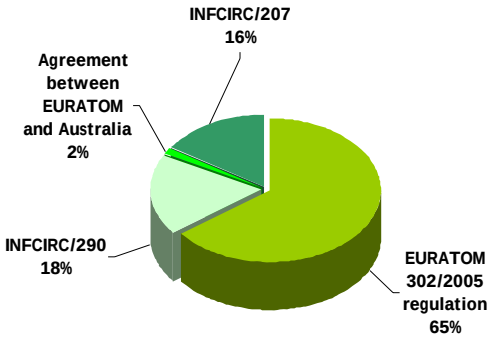
Exports notifications transmitted:

970 notifcations in 2010



Imports notifications transmitted:

769 notifications in 2010



Graphs 3 and 4: Repartition of notifications by agreement in 2010.

Consequently, a crucial need appeared to design a new information system for notifications to maintain the full compliance with the French international obligations and to enable the analysis and transmission of this huge amount of data.

3. Notification process and analysis

To ensure that its international commitments are fully met, France decided to create an integrated structure.

The operator is responsible for the content of the notification. He is the only initiator for cancellation or transfer modification (except for legal purposes) and the correspondent if the French Authorities require additional elements on the transfer.

Its notifications are transmitted to the IRSN, which makes a legal and technical analysis of the document, on behalf of the French authorities, and transfers them to the relevant authorities:

- CTE (EURATOM Technical Committee), Prime Minister Department in charge of the monitoring of the implementation of international safeguards in France.
- Ministry of foreign affairs for the French bilateral agreements.
- European Commission for the 302/2005 regulation, the agreements between EURATOM and third states and the INFCIRC/290.
- IAEA for the INFCIRC/207 (shipments to NNWS) and the INFCIRC/290 (safeguards agreement).

Picture 5 presents the notification circuit from operator to safeguards organizations and third States. The French Authorities always receive a copy of the sent notification. It can be underlined that a country can receive several notifications for the same transfer: for example Australia will receive a notification from France due to agreement [12] and one from EURATOM due to agreement [10].

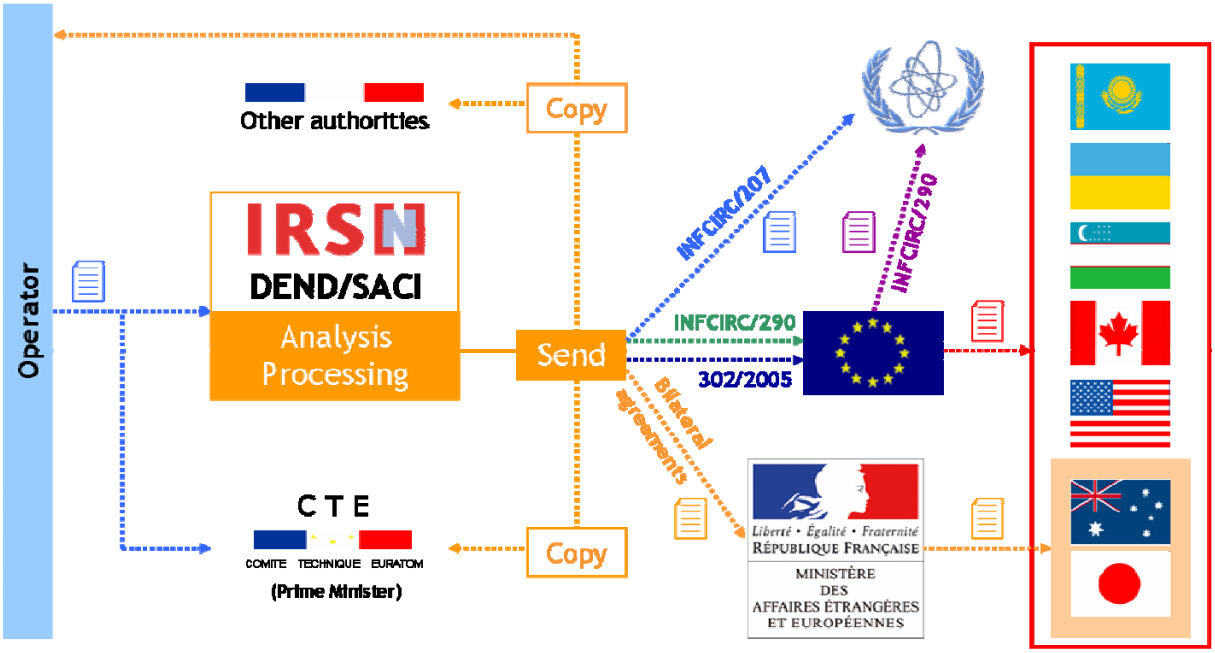


Figure 1: Notifications transmission process in France.

The analysis process consists in:

- Verifying the respect of the delays.
- Determining the relevant legal framework and the full compliance of the transfer elements (it may include the EURATOM supply Agency reference).
- Analysing if data are missing.
- Detecting errors and discrepancies.
- Transmitting data to the French Authorities and asking if necessary the operators clarification.
- Sending a uniformed notification to the relevant recipient.

This process enables French authorities to verify that international transfers of nuclear materials comply with their obligations.

4. Technical means for notification

Two means are given to the operators to notify: A website (PIMENT), operational since 2007, and a unified form, since 2004, described in the French notification manual.

4.1. The notification manual

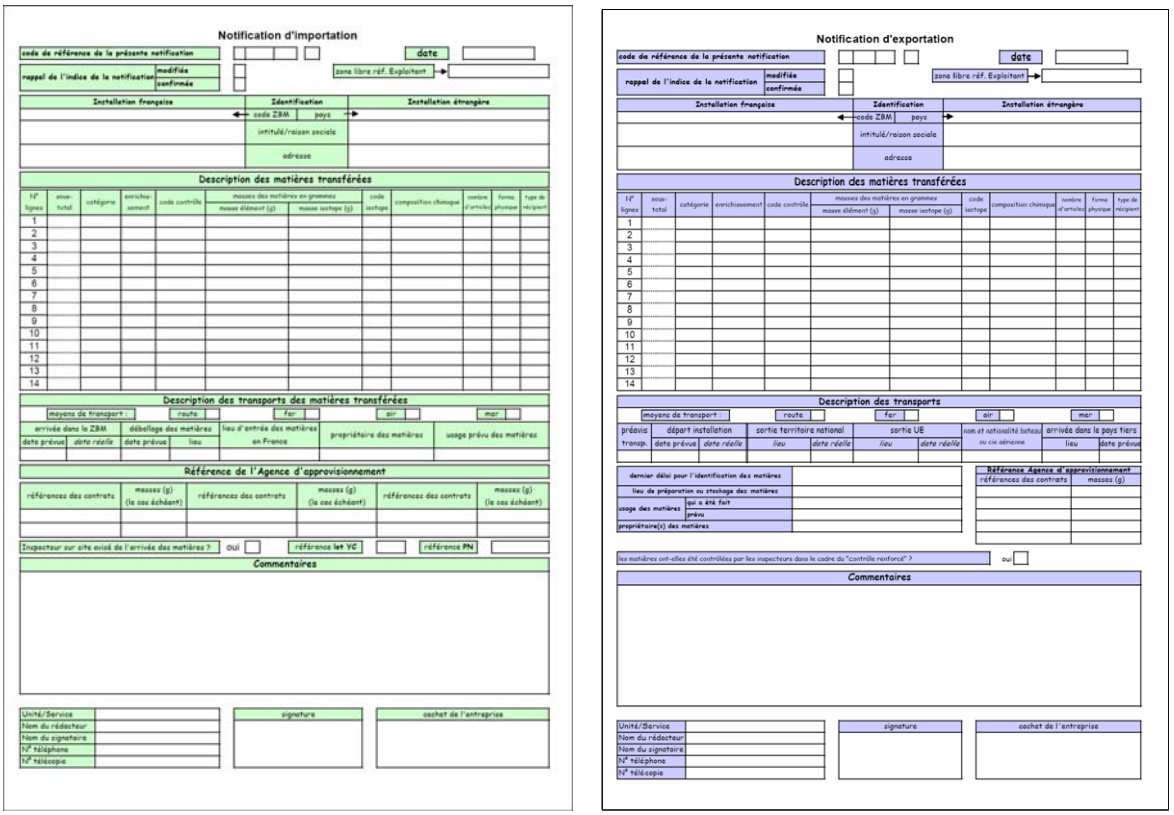
To synthesize the different regulations and to help the operators, the IRSN designed in 2010 a 108 pages comprehensive guide, which includes:

- A generic description of transfers giving the definitions, the different transfers types and the actors of the notification process.
- An exhaustive description of the regulations, including deadlines and prior consents requirements for all the regulations, the EURATOM agreements and the most frequently concerned bilateral agreements.
- A presentation of the technical means for notification, the unified form and PIMENT.
- The reference lists included in the regulations and the agreements.
- An exhaustive list of legal references.

Figure 2: Notification manual.

4.2. The unified form

The unified form was specially designed in 2005 to sum up on one sheet all the information required by agreements within an organized way, after an exhaustive examination of the legal framework. Thus, the operators can instantly give the correct information without knowing all the legal matters.



Figures 3 and 4: Unified form for imports (left) and exports (right).

4.3. PIMENT

IRSN wanted to improve and optimize the notification process introducing PIMENT, a website for the notification of nuclear materials transfers. The data are hosted in a secured zone in the IRSN head office and the connexions to the distant server are secured by a personal digital key and a SSL secured connection.

The objectives of PIMENT are:

- To improve the IRSN analysis transmission process, saving time and ensuring a high level of reliability, suppressing the recapture operation.
- To propose new features for the operators, as the previous notifications backup and access, the assistance in filling by personalizing the interface and the automate management of frequently used parameters, such as the notification reference, the foreign installations address or the EURATOM supply agency reference. The creation of notification model is also possible.
- The transmission of notifications in XML format.
- The on-line access of the French Authorities to the notifications information.

The respect of delays is a key point of the process. To address this concern, the system is designed to ensure a unavailability time in case of crash shorter than a day.

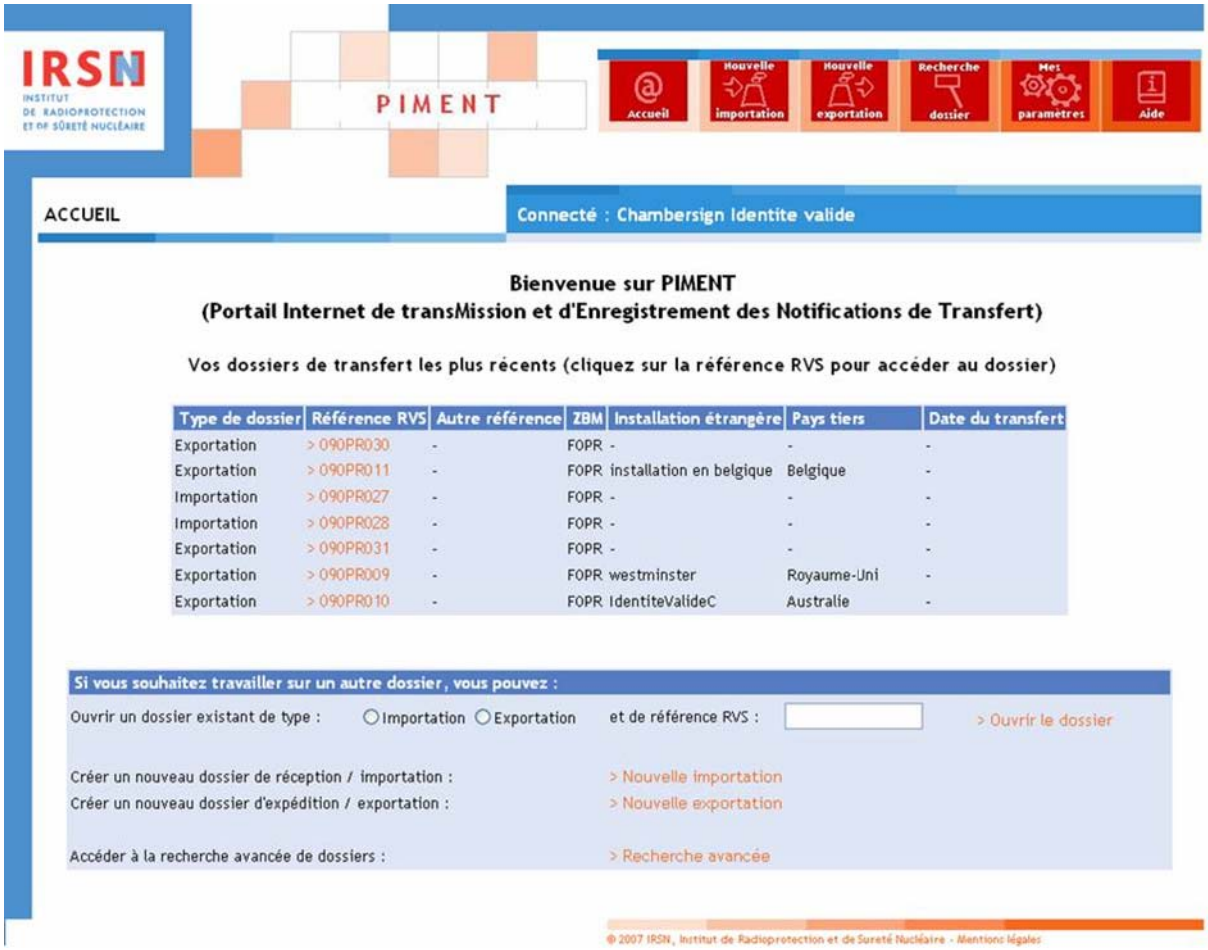
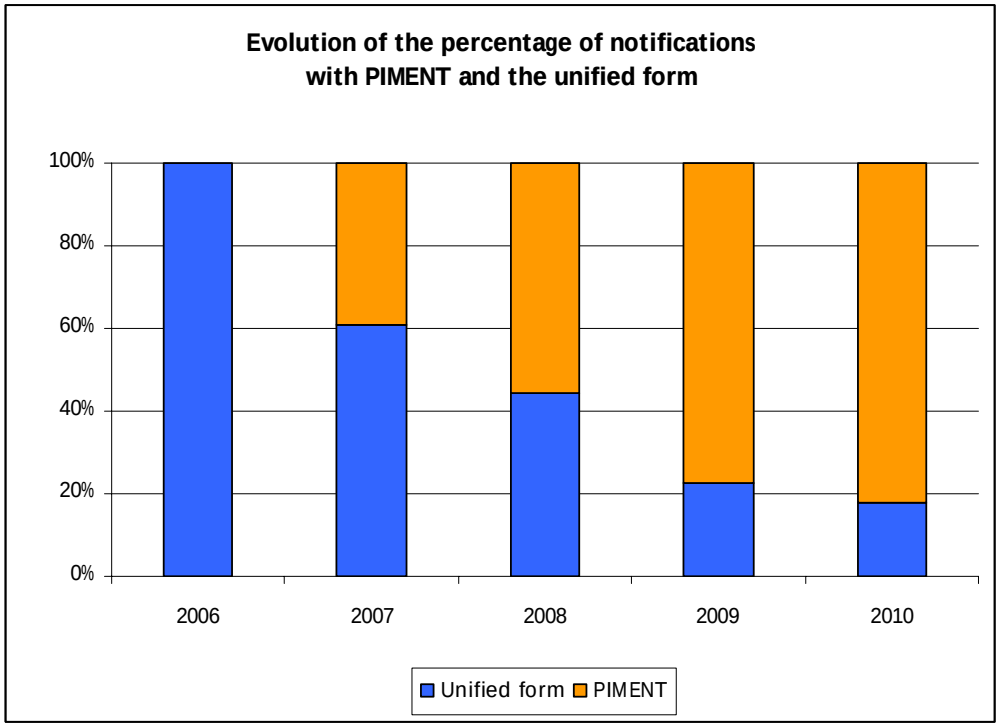


Figure 5: PIMENT user homepage.

Nowadays, the most important part of the French operators use this technical mean to notify.



Graph 5: Percentage of notifications created with PIMENT or with the unified form.

5. Advantages of the French structure for the notification process

5.1. Complying with all the regulations obligations

As we saw previously, the legal regulations and agreements edifice is complex and diverse. The advantage of an integrated system with a single focal point in charge and a unified analysis process is to earn efficiency and to ensure a systematic verification of any legal concern. It grants the insurance that France fulfils completely its obligations.

5.2. Enabling an immediate transmission of all the required elements

With the implementation of new technical means, like the website PIMENT, the analysis and transmission of data is faster. Moreover, the conception of a unified form and the introduction of constraint fields secures the filling of the notification and ensures that the document is consistent and comprehensive.

5.3. Ensuring a high level of quality

Several verifications levels are operated by France before the safeguards organization verification: It ensures a high level of confidence in the provided information. At the same time, it gives the insurance of an exhaustive transmission of all required data for each agreement and regulation, and consequently reduces the quantity of advance notifications transmitted.

5.4. Unifying the design of the notifications

The notifications always have the same aspect and information is always at the same place. Only the relevant information is transferred. These two points improve their process by the safeguards organizations.

5.5. Answering faster to the safeguards organizations questions

The analysis of the notification by the IRSN induces a better knowledge of each transfer and can induce a dialog with the operator for complementary information, which can be quicker explained or transferred to the safeguards institutions, for example unit D3 of EURATOM.

6. Acknowledgements

The authors would like to acknowledge their colleagues who have reviewed these guidelines and those who have contributed to the development and implementation of the technical notification means.

7. References

The mention “Official journal” refers to the official journal of the European Union.
The mention “JORF” refers to the Official journal of the French Republic.

[1] Treaty establishing the European Atomic Energy Community (consolidated version 2010/C 84/01).

[2] Commission regulation (EURATOM) No 302/2005 of 8 February 2005 on the application of EURATOM safeguards.

[3] Notification to the Agency of exports and imports of nuclear material (INFCIRC/207/add.1) – March 1984.

[4] Agreement of 27 July 1978 between France, the European Atomic Energy Community the European Atomic Energy Community and the International Atomic Energy Agency for the application of safeguards in France (INFCIRC/290).

[5] Agreement for cooperation in the peaceful uses of nuclear energy between the European Atomic Energy Community and the United States of America (Official Journal 017, 19/03/1959 P.0312).

[6] Agreement between the Government of Canada and the European Atomic Energy Community (EURATOM) for cooperation in the peaceful uses of atomic energy (Official Journal 060, 24/11/1959 P.1165 – 1176).

[7] Agreement between the Government of Japan and the European Atomic Energy Community for co-operation in the peaceful uses of nuclear energy (Official Journal L 032, 06/02/2007 P.0065 – 0075 and Official Journal L 219, 24/08/2007 P.0100 – 0110).

[8] Agreement for cooperation in the peaceful uses of nuclear energy between the European Atomic Energy Community (EURATOM) and the Government of the Republic of Uzbekistan (Official Journal L 269, 21/10/2003 P.0009 – 0017).

[9] Agreement between the European Atomic Energy Community and the Cabinet of Ministers of Ukraine for Co-operation in the Peaceful Uses of Nuclear Energy (Official Journal L 261, 22/09/2006 P.0026 – 0031).

[10] Agreement for co-operation in the peaceful uses of nuclear energy between the European Atomic Energy Community and the Government of the Republic of Kazakhstan (Official Journal L 010, 15/01/2009 P.0016 – 0021).

[11] Agreement between the Government of Australia and the European Atomic Energy Community concerning transfers of nuclear material from Australia to the European Atomic Energy Community (Official Journal L 281, 04/10/1982 P.0008 – 0020).

[11] Agreement of 7 January 1981 between the Government of France and the Government of Australia concerning the transfers between France and Australia (JORF 05/03/1985 P.2712-2716).

[12] The structure and content of agreements between the Agency and states required in connection with the treaty on the non-proliferation of nuclear weapons (INFCIRC/153).

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Analysis of nuclear materials in environmental samples for safeguards purposes before and during the decommissioning of nuclear facilities

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Abstract:

A comprehensive environmental monitoring and analysis of nuclear materials (especially U and Pu) in environmental samples for safeguards purposes is essential prior to, during and following the decommissioning of nuclear facilities. Measuring nuclear signatures contributes to the safeguards assessment of the process and to the detection of undeclared activities. The evaluation of measurement data should be based on reference data (base line) obtained prior to the start of decommissioning which process theoretically can be used for cover undeclared activities.

In Hungary no experience has yet been accumulated as nuclear facilities (4 NPP units and 1 research and 1 training reactor) will remain in operation foreseeable for more than 10 years. However, environmental sampling and monitoring should start prior to decommissioning as prescribed in international recommendations and the EU directive.

Environmental monitoring of radioactive contamination is performed in Hungary both by the authorities and nuclear facilities in order to control radioactive discharge and assess its impact on the environment and the public. Uranium and other actinides – essential for safeguards purposes – are usually not analyzed in these samples.

The development for the analysis of U and Pu content and isotope ratios in environmental samples, collected in the vicinity of Hungarian nuclear facilities has been started at the Institute of Isotopes. Samples have been collected using the sampling locations of the environmental monitoring systems of the Hungarian nuclear facilities and the analysis of the samples was carried out by ICP-SFMS technique. The results of these analyses complemented with continuing sampling will form the reference baseline for uranium and plutonium for safeguards purposes in the area of nuclear facilities.

Keywords: environmental sampling; decommissioning, safeguards, ICP-SFMS

1. Introduction

In case of taking a decision of closing a nuclear power plant permanently, the facility must be decommissioned by safely removing it from service and reducing residual radioactivity.
The decommissioning of a nuclear power plant contains the cleanup of radioactively contaminated plant systems and structures and the removing of the radioactive fuel [1].

One of the basic criteria of the decommissioning is the environmental monitoring and so-called environmental impact analysis or assessment which is obligatory during the decommissioning procedure and even required to obtain a decommissioning licence [2]. The necessary decommissioning environmental monitoring must be completed to ensure the detection and decreasing of off-site impacts caused by radioactive or hazardous materials released from decommissioning activities at nuclear facilities [3].

Additionally, the environmental impact study has to consist of a baseline related to the condition prior to the decommissioning and the environmental report will also describe the likely environmental effects that may arise from undertaking decommissioning and site clean up in the future.

The nuclear sites to be decommissioned should have a documented environmental baseline prior to the start of operations. These data should be reviewed, and the sample points used to establish the initial background level should be utilized through subsequent environmental reports to update the background value of these locations before decommissioning operations begin. This documentation should be completed by additional samples taken from the original baseline sampling locations and new locations for pathways resulting from decommissioning activities. The environmental or radiation protection programs usually maintain environmental sampling locations that can provide baseline information for air radioactivity, radon levels, external beta/gamma radiation values, or trace element levels in ambient air [4]. Besides analysis of soil, plant and some food samples in the aspect of radiation protection is also required.

The international standards and guidelines (IAEA, ASTM, NRC, NDA) describe the requirements and implementation of the environmental studies and monitoring but these take into account only radiation protection and decontamination aspects [1-4]. Safeguards purpose of the environmental monitoring could be supported to the Wide Area Environmental Sampling of IAEA [5, 6] as well as could help the safeguards verification and control of the sites under decommissioning.

The aim of this paper was to present the results of the preliminary study on the analysis of environmental samples for the purpose of safeguards aspects in the environmental monitoring during the decommissioning of nuclear facilities. For this aim environmental samples were collect and analyze close to the nuclear installations in Hungary using the sampling locations of their present monitoring systems and to start the establishment of the baseline before the first decommissioning in the country.

2. Experimental

2.1. Instrumentation

Measurements of environmental samples were carried out using a double focusing magnetic sector inductively coupled plasma mass spectrometer (ICP-SFMS) equipped with a single electron multiplier (ELEMENT2, Thermo Electron Corp., Germany). Prior to the sample analysis the instrument was tuned using a 1 ng g^{-1} multielemental solution (Merck, Darmstadt, Germany). The optimization was carried out with respect to maximum uranium sensitivity and low UO^+/U^+ ratio. The sensitivity was approximately 1×10^6 cps for 1 ng g^{-1} ^{238}U.

All measurements were carried out in low resolution mode ($m/\Delta m$= 300) using a Teflon nebulizer in self aspirating mode (flow rate was approximately 100 µl min^{-1}) in combination with a desolvation unit (Aridus, CETAC Technologies Inc., USA) for Pu measurements that removes most of the solvent thus significantly decreases hydride and oxide interferences. A combined spray chamber (Scott – cyclone) was used for analysis of U that ensures the precise isotope ratio measurement with low uncertainty.

2.2. Samples

12 samples (7 soil, 4 plant (grass) and 1 sludge from Danube) were collected and analyzed by ICP-SFMS. Samples originated from sampling points of the monitoring systems of the nuclear reactors (KFKI Research Reactor and Training Reactor at the Institute of Nuclear Techniques of the Budapest University of Technology and Economics) and the PAKS Nuclear Power Plant (NPP) in Hungary. The sample collection was positioned close to the reactors and isotope storage at KFKI Campus. Samples were dried and homogenized by the Environmental Monitoring Laboratory of Paks NPP.
Figure 1 and Figure 2 show the sampling points at KFKI Campus and NPP. Sampling points are indicated using numbers in the map and essential ‘hot’ points are also presented in Figure 1.

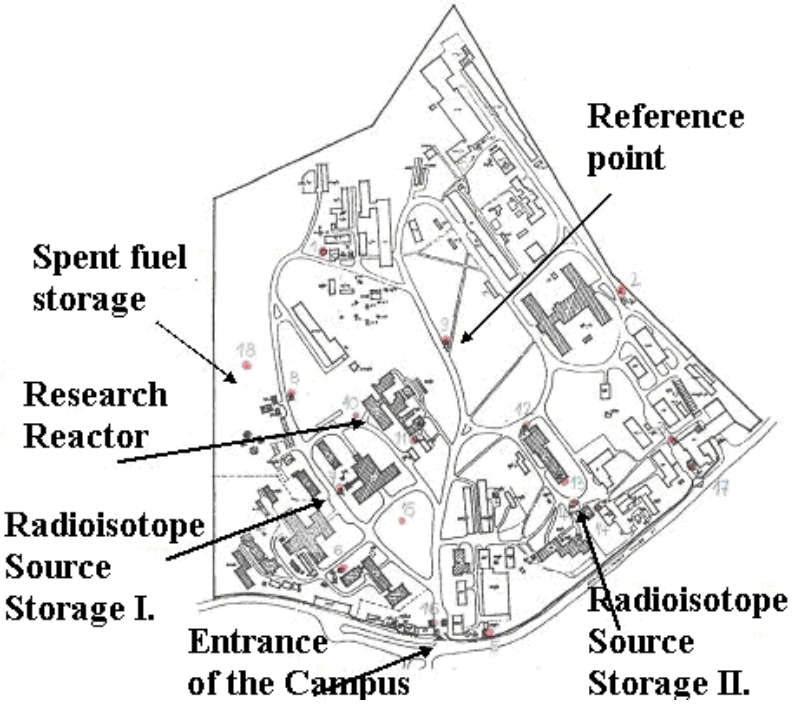

Figure 1: Map of the KFKI Campus and the sampling points

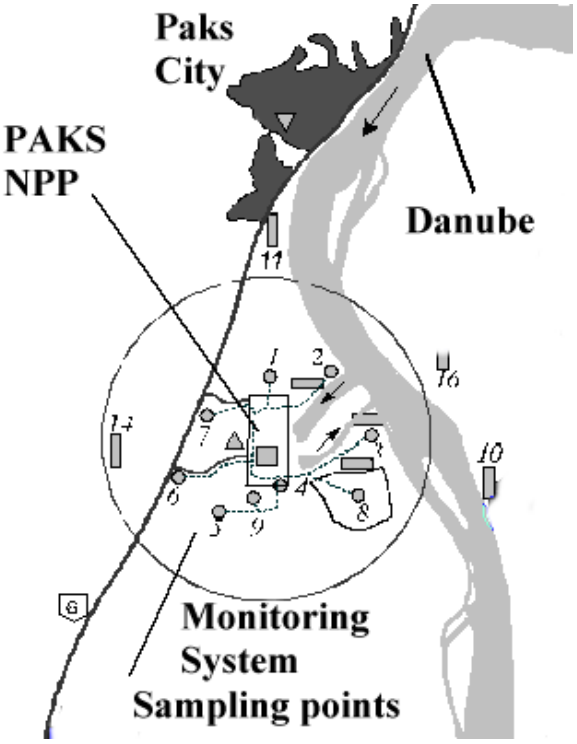

Figure 2: Map of the Paks NPP and the sampling points of the environmental monitoring system

The whole analytical procedure was checked using a reference material (IAEA-385, Irish Sea sediment).

2.3. Sample preparation

Approximately, 3-5 g homogenized samples were weighted into a glass beaker and 400 µl of ^{242}Pu and 200 µl ^{233}U of tracers were added to the samples by weighing. Following the addition of 10 ml concentrated HNO_3, the samples were digested using hot plate. The covered samples were heated up to 120 °C 24 hours long and 2-3 ml H_2O_2 was added to the samples time to time to digest the organic components. After digestion the samples were evaporated to reduce the HNO_3 volume up to 5 ml. The samples were filtered gravimetrically through a double Whatman 2 type paper filter into a 50 ml polyethylene centrifuge tube. The samples were diluted to approximately 40 ml. To the filtered sample 1 ml of 0.1 g ml^{-1} $Ca(NO_3)_2$ solution was added followed by the careful addition of 300 mg of $NH_2OH.HCl$. After complete dissolution the tube was placed in a 90 °C water bath for 10 minutes. During this step plutonium is reduced to Pu(III). After cooling 8 ml of concentrated HF was added carefully to the sample and mixed thoroughly. After 15 minutes the precipitate was centrifuged and the supernatant was discarded. Supernatant was used for determination of uranium content in the samples.

The precipitate was dissolved in 15 ml 3M HNO_3 and 500 mg of H_3BO_3 by placing in a hot water bath and vigorous shaking. The plutonium was separated by extraction chromatography using TEVA® resin. After conditioning of the column with 10 ml of 3M HNO_3 the sample was loaded on the column. After loading the tube and the column were rinsed twice with 2.5 ml of 3M HNO_3. The retained U and Th were eluted with 20 ml of 3M HNO_3 and 15 ml 6M HCl, respectively. Finally, plutonium was eluted by 20 ml of 0.1M HNO_3/0.1M HF collected in a PFA beaker. The solution was repeatedly evaporated to almost dryness (3 times) with successive addition of 2 ml of ultrapure HNO_3 to remove HF and organic content. The residue was dissolved in 1 w/w% distilled nitric acid (1.6 ml) with gentle heating on a hotplate and was measured the Pu in the sample by ICP-SFMS [7].

The supernatant was evaporated till 0.1 ml and was dissolved in 5 ml 8 M HNO_3. Uranium was separated by extraction chromatography using UTEVA® resin. After conditioning of the column with 20 ml of high purity Milli-Q water and 20 ml 8 M HNO_3 the sample was loaded on the column. After loading the tube and the column were rinsed twice with 2.5 ml of 8M HNO_3. The matrix components of the sample were eluted with 20 ml of 8M HNO_3. Finally, uranium was eluted by 20 ml of 0.1M HCl collected in a PFA beaker. The solution was repeatedly evaporated to almost dryness (3 times) with successive addition of 2 ml of ultrapure HNO_3. The residue was dissolved in 1 w/w% distilled nitric acid (1.6 ml) with gentle heating on a hotplate and was measured the Pu in the sample by ICP-SFMS.

3. Results and discussion

The collected and prepared environmental samples were analyzed by ICP-SFMS instrument. Table 1 and Table 2 illustrate the isotopic composition and concentration results obtained for uranium and plutonium.

Origin of the sample	Sample I.D.	Sample type	^{235}U atom%	^{234}U atom%	Total U content µg g^{-1}
Training Reactor (INT, BUTE)	0746	soil	0.7359 ± 0.072	0.0058 ± 0.001	3.00 ± 0.33
	0747	plant	0.7607 ± 0.018	0.0047 ± 0.0001	0.471 ± 0.05
Research Reactor (KFKI Campus)	0809	soil	0.7711 ± 0.074	0.0051 ± 0.001	2.51 ± 0.23
	0810	plant	0.7442 ± 0.032	0.0067 ± 0.0003	0.493 ± 0.05
	0815	soil	0.7161 ± 0.051	0.0054 ± 0.0007	7.27 ± 0.71
	0816	plant	0.8091 ± 0.055	0.0065 ± 0.0004	0.413 ± 0.04
	0817	soil	0.6739 ± 0.005	0.0045 ± 0.0004	1.72 ± 0.17
	0818	plant	0.7525 ± 0.055	0.0065 ± 0.0005	0.081 ± 0.008
Paks NPP	0839	sludge	0.7260 ± 0.042	0.0053 ± 0.0006	0.391 ± 0.04
	0852	soil	0.7488 ± 0.011	0.0059 ± 0.0001	5.29 ± 0.49
	0855	soil	0.7080 ± 0.029	0.0061 ± 0.0002	1.34 ± 0.13
	0858	soil	0.7665 ± 0.01	0.0062 ± 0.0001	2.72 ± 0.27

Table 1: Uranium isotopic composition and total uranium content in the environmental samples (k=2)

Origin of the sample	Sample I.D.	Sample type	$^{240}Pu/^{239}Pu$ atomic ratio	^{239}Pu conc. (fg g^{-1})	^{240}Pu conc. (fg g^{-1})
Training Reactor (INT, BUTE)	0746	soil	0.20 ± 0.02	73.8 ± 8.86	18.8 ± 2.54
	0747	plant	*n.d. (< 0.0215)	*n.d. (< 0.83)	*n.d. (< 0.44)
Research Reactor (KFKI Campus)	0809	soil	0.206 ± 0.02	37.5 ± 4.56	7.74 ± 0.92
	0810	plant	0.145 ± 0.01	42.3 ± 4.98	6.15 ± 0.76
	0815	soil	0.192 ± 0.019	32.5 ± 4.12	6.25 ± 0.77
	0816	plant	0.185 ± 0.02	35.4 ± 3.99	3.52 ± 0.43
	0817	soil	0.216 ± 0.02	23 ± 3.78	5.98 ± 0.66
	0818	plant	0.121 ± 0.02	26.6 ± 3.01	3.23 ± 0.35
Paks NPP	0839	sludge	0.128 ± 0.02	34.7 ± 2.98	4.45 ± 0.53
	0852	soil	0.173 ± 0.02	22.3 ± 2.42	1.62 ± 0.21
	0855	soil	0.191 ± 0.01	17.4 ± 2.11	1.57 ± 0.26
	0858	soil	0.184 ± 0.02	26.9 ± 2.75	4.96 ± 0.55

* n.d.: Not detectable

Table 2: Plutonium isotopic composition and plutonium isotopic (^{239}Pu, ^{240}Pu) content in the environmental samples (k=2)

The uranium atomic percent values show the natural uranium composition in the analyzed environmental samples. ^{236}U was below the detection limits in all cases and the total uranium content was in the range of normal natural uranium level. Therefore, it could be concluded that uranium contamination originated from nuclear activity was not found in the environmental samples..

Plutonium results obtained show the typical isotopic ratio and amount originated from global fall-out. ^{241}Pu was not detected in the samples (below detection limits). The reactor or weapon origin plutonium isotope ratios have not been observed. Plutonium contamination was not determined in the environment of the studied reactors and NPP.

Plutonium was below a detection limit in the plant sample originated from the environment of the Training Reactor. Presumably, higher sample amount is necessary for further investigations.

Measurements were validated using the IAEA-385 reference material. Results obtained can be seen in Table 3 and Table 4. Analytical results agreed with the reference values within the combined uncertainties.

Sample	^{238}U conc. (ng g^{-1})	^{235}U conc. (ng g^{-1})	^{234}U conc. (ng g^{-1})
IAEA-385 meas.	2314 ± 198	17.02 ± 1.09	0.119 ± 0.009
IAEA-385 ref.[8]	2364 (2251-2453)	17.0 (15.5-18.9)	0.114 (0.109-0.121)

Table 3: Results of uranium isotopic measurements in the testing of the analytical procedure using IAEA-385 reference material (k=2)

Sample	^{239}Pu conc. (fg g^{-1})	^{240}Pu conc. (fg g^{-1})	^{241}Pu conc. (fg g^{-1})
IAEA-385 meas.	799 ± 75	142 ± 31	< 5.5
IAEA-385 ref.[8]	836 (566-901)	140 (115-157)	-

Table 4: Results of plutonium isotopic measurements in the testing of the analytical procedure using IAEA-385 reference material (k=2)

4. Conclusion

Establishing the reference database and environmental monitoring system for safeguards purposes is recommended before and during the decommissioning of nuclear facilities.

This paper is a preliminary study for planning and feasibility of environmental monitoring program for safeguards purposes. The monitoring system should contain an extended baseline study and regular control measurements.

For the achievement of this aim and test of the possible system, environmental samples were collected, prepared and analyzed using the sampling points of the present environmental monitoring systems of the nuclear facilities in Hungary. The future monitoring system should be based on the present monitoring systems to be completed it with other sampling points and aspects after an exhaustive planning.

On the basis of the results obtained, it could be concluded that in the environment of the Hungarian nuclear facilities neither uranium nor plutonium was found which could originate from local nuclear activities. Uranium was found in natural composition and amount and plutonium was found in the typical isotopic ratios and amount of the global fall-out. Besides, these measurements give data for a reference database. For the creation of the complete database more samples should be collected and analyzed.

5. Acknowledgement

The authors wish to thank the co-workers of PA Zrt., especially László Manga, the colleagues of the Institute of Nuclear Techniques of the Budapest University of Technology and Economics and Dr. László Sági (head of the Environmental Monitoring System at KFKI Campus) for their help and for the samples. The Hungarian Atomic Energy Authority deserves thanks for financial support. Dr. Tamás Bíró's efforts and scientific advices were also highly appreciated.

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Safeguards Implementation during the BN-350 Fast Breeder Reactor Spent Fuel Transfer

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Abstract:

The BN-350 Nuclear Power Plant is a sodium-cooled Fast Breeder Reactor located in Aktau (Kazakhstan), which was in operation from 1973 to 1998 and has been definitively shut-down since 1999. The reactor was designed to use LEU, HEU and MOX fuel. Spent fuel was conditioned in steel containers that were welded shut.

All of the containers have been loaded into reinforced concrete Dual Use Casks (DUC), that were temporarily stored at the Dry Spent Fuel Storage co-located with the BN-350 reactor, before being transferred to a long-term storage at Baikal-1, located in north-eastern Kazakhstan (Semipalatinsk region), over 3000 km away. The transfer was completed in November 2010.

This paper discusses the safeguards approach implemented to maintain the continuity of knowledge during the spent fuel transfer campaign.

Keywords: spent fuel, transportation, storage, safeguards

1. Introduction

The BN-350 Nuclear Power Plant, in operation from 1973 to 1998, served multiple purposes: to breed plutonium and to operate for electricity generation, district heating, and water desalinization for the city of Aktau. The BN-350 reactor was the only power reactor in the world used for industrial scale desalinization (up to 100,000 tonnes/day).

The reactor was designed to use uranium fuel enriched to 17%, 21% and 26% in ^{235}U, as well as MOX fuel (23% Pu). The core was surrounded by blanket assemblies containing depleted uranium for Pu breeding. The BN-350 was capable of breeding over 110 kg of plutonium each year.

Irradiated assemblies were routinely returned to Russia's Mayak Production Association (PO Mayak) in Chelyabinsk Oblast for reprocessing after cooling in the BN-350 pond. After the dissolution of the Soviet Union in 1991 spent fuel shipments to Russia stopped. Many of the fuel assemblies irradiated over the 25 years of operation (about 3000 items, containing slightly less than 3 tonnes Pu) remained in the reactor facility's cooling pond [1] (see Fig. 1).

Within the framework of the BN-350 Spent Fuel Disposition Project, funded by USDOE and Kazakhstan, from 1999 to 2001 all the spent fuel assemblies (SFA) were verified by partial defect test and canned in 479 sealed steel canisters (referred to as “canisters” in the following), each containing 4 or 6 SFAs, which have been stored in the spent fuel pond (SFP) and kept under dual containment and surveillance (C/S) based on seals and surveillance until 2008, when the next active phase of the project started [2].

A specific under water instrument (Spent Fuel Coincidence Counter, SFCC) was designed for the verification of these SFAs, which were all measured for partial defect to assess the quantity of Pu. Each assembly was measured under water at 5 points along its vertical axis.

A dedicated surveillance system (Multi Instrument Monitoring System, MIMS) was designed to maintain the continuity of knowledge of this highly safeguards-sensitive nuclear material. This was achieved with the support of USDOE through Los Alamos National Laboratory and other laboratories in the US. The system includes radiation detection monitoring capabilities (gamma and neutron detectors) which are integrated with video surveillance [3].

The continuity of knowledge (CoK) of the nuclear material has been successfully maintained since the end of the first canning campaign in 2001.

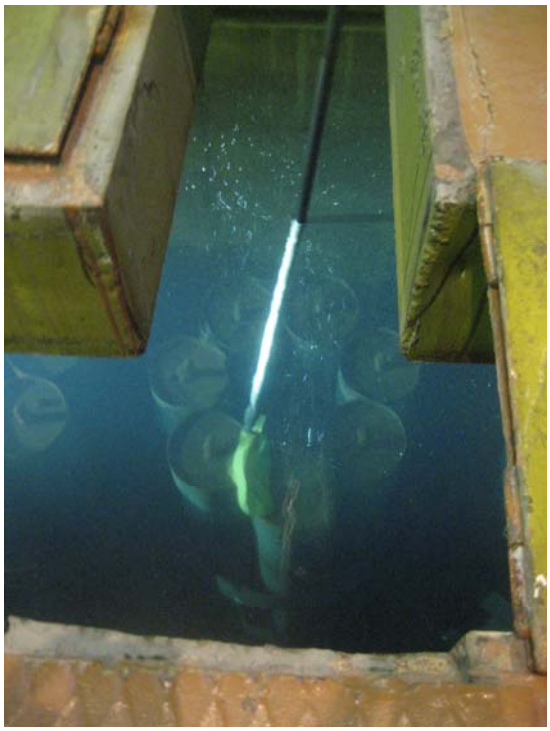

Fig. 1 – Canisters in the SFP

Fig. 2 – A DUC at the DSFS

2. The facilities

The project was resumed in 2006, when the design and construction of the different facilities connected to the project entered an active phase. Reinforced concrete Dual Use Casks (DUC) were designed to be used both for transportation and storage of the canisters. Each one could host 8 canisters (see Fig. 2).

An interim Dry Spent Fuel Storage (DSFS), co-located with the reactor facility in Aktau and surrounded by an additional fence, was built to store 60 DUCs. The storage facility became operational in January 2009, when the first DUC was stored on the concrete pad.

A long term DSFS, located in the Semipalatinsk region (north-eastern Kazakhstan), was built to store 60 DUCs for 50 years. The storage facility became operational in February 2010, when the first DUC was stored on the concrete pad.

The facility consists of two functional parts, one (the receiving area) located near the railway station at Kurchatov City, the other (the DSFS itself) at the Baikal site, about 70 km away from the above mentioned station. The receiving area is equipped with a crane (150 t) to unload each DUC together with

its protective over-pack from the railcar and load it onto a specially designed truck. The protective over-packs had to be removed after delivery to the DSFS to be shipped back to Aktau.

3. The spent fuel transfer process: packaging and transfer to the interim DSFS

The entire population of canisters has been loaded into DUCs using the hot cell at the BN-350 facility, and then transferred to the interim DSFS. The loading of the first DUC started in November 2008; the last DUC was transferred out of the BN-350 facility to the interim DSFS and loaded onto the train to be transported to the long-term DSFS in November 2010.

A safeguards approach has been developed according to safeguards criteria in order to ensure that the CoK is maintained through the transfer of the canisters from the pond in the BN-350 reactor building, where they were under a dual C/S system, to the interim DSFS. Similarly, the nuclear material was maintained under a C/S system with multiple backup during the transfer activities and finally under a dual C/S system at the interim DSFS.

The transfer process encompassed all steps necessary to package the canisters into the DUCs and to transfer the DUCs to the interim DSFS. The canisters were transferred under water into the hot cell, where their transit was detected by two underwater gamma detectors, then washed and dried in air and inserted into shielded positions, where they were tested for leakage. Two gamma and neutron detectors located at opposite ends of the hot cell were able to detect the presence of spent fuel in air and the direction of its movement. After being tested for leakage, the canisters were inserted into the DUC, placed in the rail corridor underneath the hot cell, where another gamma and neutron detector provided the information about this latter movement of the spent fuel.

Once the DUC was filled with 8 canisters, it was closed with three lids providing additional biological shielding and physical protection. The final step of the process was the transfer by rail to the interim DSFS.

3.1. The safeguards approach

The following diversion scenarios had to be taken into account during the packaging and transfer of the nuclear material to the interim DSFS:

- The substitution of one or several full canisters by others containing dummy elements during the loading period (before sealing the DUC).

- The substitution of a full DUC with an empty one or with one filled with dummies or by one partially filled. However, re-opening of a DUC to remove a canister would require its transportation to a hot cell.

The hot cell of the facility is the only known place on the routine spent fuel transfer route to the dry storage where canisters could be opened (and/or fuel assemblies could be dismantled or cut). The canister drying and testing, the possible re-opening and replacement of the containment (canister), and subsequent re-welding would take place in the hot cell.

After the 8 canisters have been loaded into the DUC, upon its transfer to the dry storage facility and while on storage, the diversion assumption was the removal of one or several canisters for clandestine opening and unloading after a clandestine removal of a DUC. For concealment, a partially loaded DUC might have been returned to the DSFS.

The C/S system during the transfer was based on seals, surveillance using radiation monitors and optical surveillance, and human surveillance. In order to reduce the possibility of failure, multiple independent backup systems were installed. The time period between the cutting of the seals on any canister in the SFP and the application of dual C/S at the interim storage site was always less than the timeliness period (3 months + 3 weeks).

4. The spent fuel transfer process: transfer to the long-term DSFS

The entire population of the 60 DUCs was transferred to the long-term DSFS in Baikal-1: 12 transportations with 5 DUCs each took place. The activity was started in February 2010 and completed in November 2010, when the last DUC was immobilized at the storage site (see Fig. 3).

The transfer process included all steps necessary to transfer the DUC's from the interim to the long-term DSFS. Each DUC was placed into its protective over-pack at the interim storage in Aktau, then loaded onto the railcar. The train transported 5 DUCs per trip, about 3000 km away through Kazakhstan.

Upon arrival in the receiving area at the Kurchatov City railway station, the DUCs were transferred from the railcar to a specially designed truck and transported to Baikal-1.

Upon arrival at Baikal-1, the DUCs were unloaded from the truck, the over-pack was removed and the DUC was installed in its final position on the concrete pad.

4.1. The safeguards approach

The following diversion scenarios had to be taken into account during the transfer of the nuclear material to the long-term DSFS:

- Opening of a DUC to remove one or more canisters; this would require its transportation to a hot cell.
- The substitution of a full DUC with an empty one or with one filled with dummies or by one partially filled.

The hot cell at the BN-350 facility is the only known place on the routine spent fuel transfer route to the long-term dry storage where the casks could be opened.

During the DUC transfer to the dry storage facility and while on storage, the diversion assumption is the removal of one or several canisters for clandestine opening and unloading after a clandestine removal of a DUC. For concealment, a partially loaded DUC might have been returned to the DSFS.

The C/S system during the transfer from Aktau to Kurchatov was based on seals, while during the transfer from Kurchatov to Baikal it was based on seals and/or human surveillance. The time period between removing a DUC from the dual C/S at the interim DSFS in Aktau and the application of dual C/S at the long-term DSFS in Baikal-1 was always less than the timeliness period (3 months + 3 weeks).

Nuclear material will be stored at the long term DSFS at Baikal-1, which has been designed for a period of 50 years.

Fig. 3 – The insert for immobilizing seal

Fig. 4 – The transport wagon

5. The safeguards system at the interim and long-term DSFS

The dual C/S system at both dry storages consists of the storage cask containment and seals, including the immobilizing seals. In particular it consists of the following two functionally independent components that are not subject to any common tampering or failure mode:

- Fiber-optical wire using a COBRA seal attached to at least two of the DUC containment closure mechanisms;

- Metallic wire using an E-type seal attached to at least two of the DUC containment closure mechanisms, which are different from the previous ones.

The bolts on the top of the DUCs are considered as the containment closure mechanisms mentioned above.

Each DUC is also immobilized by sealing it to the ground with an E-type or fiber-optical seal attached to a bolt that is screwed onto the side of the cask.

For the immobilization to the ground metallic anchors were welded to the pad reinforcing rebars (see Fig. 4) and embedded in the storage pad concrete. The anchors have such a shape that, if removed, a visible portion of the concrete will be damaged. Special IAEA bolts (Stud Sealing Device) were screwed into the holes in the anchor and on the side of the DUC. The seal wire is mechanically protected against accidental damage with aluminum conduits.

Should the CoK of the nuclear material be lost, due to seals failure or other contingencies, a re-verification method will be needed in order to re-establish the knowledge of the material without the need of opening the DUC. A method based on neutron measurements on the outer surface of a DUC has been developed by Los Alamos National Laboratory and is being evaluated at the Agency to be approved for inspection use. The scope of this method is to “fingerprint” each DUC and detect at any time the removal of one canister from within the DUC. All 60 DUCs have been measured before leaving the interim DSFS, in order to provide an initial set of data for the evaluation of the method [4].

6. Conclusions

The whole population of spent fuel present at the BN-350 reactor was measured to assess the Pu content and packaged into sealed stainless steel containers during the first phase of the BN-350 Spent Fuel Disposition Project (1999-2001). From 2008 to 2010 these canisters were loaded into reinforced concrete Dual Use Casks (DUC) and transported 3000 km away through Kazakhstan. The continuity of knowledge was successfully maintained throughout the whole process with a combination of radiation detectors, optical surveillance, seals and human surveillance. The whole population of 60 DUCs is now stored in the Semipalatinsk region under a dual Containment and Surveillance system based on seals.

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Object-based Change Detection Using Very High-resolution Satellite Data

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Abstract:

With the advances in satellite sensor technologies as to spatial resolution, the concept of object-based image analysis (OBIA) has become widely-used in different remote sensing applications and involved the development of some commercial off-the-shelf systems. None of the systems available today is able to achieve the accuracy of human image interpreters; however, all of them are able to at least provide a pre-selection of relevant object features and image objects. The procedures implemented in the systems still show some shortcomings in terms of segmentation, feature extraction, multitemporal analysis, and others. Against this background, this paper aims to present some recent developments and advances in object-based change detection for nuclear safeguards applications. In detail, procedures based on Multiresolution Segmentation (MRS) and Multivariate Alteration Detection (MAD) were implemented as C++ stand-alone programmes and Definiens Developer plug-in.

The paper introduces the methodologies, describes the implementation and gives some (experimental) results on nuclear safeguards applications using simulated and real data.

Keywords: change detection; segmentation; multivariate alteration detection; object-based image analysis

1. Introduction

Since the discovery of the Iraqi nuclear weapons programme in 1990, satellite imagery has become a powerful and intensely used tool for verifying the States' compliance with their NPT safeguards agreements. Satellite imagery can assist in the evaluation of site declarations, the detection of undeclared nuclear facilities, and the preparation of inspections or other visits. For analysing and assessing the development of sites under construction (whether declared or not) and for monitoring clandestine facilities, multitemporal satellite imagery acquired over the same area at different times is needed. In remote sensing processing, to the comparison of two or more images is known as change detection. However, when it comes to the automation of change detection procedures, several shortcomings have to be considered, in particular when very high-resolution satellite imagery is being used. Seasonal changes of vegetation, different shadow directions, different acquisition angles and poor co-registration might cause many false alarm changes that need to be eliminated interactively by the user. On the other hand, it is a repetitive and exhausting task for image analysts to find relevant changes in large imagery only by visual interpretation.

Nevertheless, a huge number of data processing methods trying to automate change detection were proposed (see e.g. [6]). Methods analysing difference images [7], classification based approaches [8] and kernel based methods such as principal component analysis [9] or multivariate alteration detection (MAD) [2], to name only a few examples. In this paper we will focus on the method of iteratively re-weighted multivariate alteration detection (IR-MAD) proposed by Nielsen [2].
Pixel-based approaches generally compare the spectral values of corresponding pixels of different acquisition times. However, as the spatial resolution of remote imaging sensors has increased, a single pixel's area on the ground has decreased. Thus, aggregating spatially connected pixels into

homogeneous objects, also referred to as segmentation, has become more and more important. This paradigm is called object-based image analysis [10].

Change detection can also be carried out based on the image objects. Earlier studies [4, 5] presented a method which adapts the use of object-based image analysis to the task of change detection. The flow chart in Figure 1 outlines the procedure.

Fig. 1: Object-based change detection procedure.

The process started with the pre-processing step consisting of co-registration and radiometric normalisation. Then, the aggregation of the pixels, the so-called segmentation, followed and provided image objects as the basis for the further analysis. In the third step, relevant object features were selected from a variety of spectral, textural, relational and shape features. Feature extraction was followed by a linear transformation of the feature space using the method of multivariate alteration detection which enhances the change information in the difference image. In the fifth step of the process, a clustering of the objects was carried out in the transformed feature space using the fuzzy maximum likelihood (FMLE) method. Finally, a post-processing step was applied in order to correct errors during clustering using a label relaxation technique. For a more detailed discussion of the change detection methodology see [4].

One of the crucial steps in this procedure is the segmentation step. In principle, three different ways of segmentation could be used for object-based change detection:

1. Segment both images separately.
2. Segment the first image and assign this segmentation to the second image.
3. Segment both images as a stack.

Each of these methods has severe drawbacks concerning the use of shape features, the robustness as well as the quality of the segmentation results or the problem of linking corresponding objects between the two acquisition times (see [3] for a comprehensive discussion).

Therefore, we will present a new way of segmentation for object-based change detection. The method is based on the Multiresolution Segmentation (MRS) algorithm which is a core component of the eCognition software [11]. It aims to produce similar segments in image regions without actual changes between the acquisition times and to provide different segments for changed image regions. Furthermore, we will show how to integrate the segmentations into the change detection process.

A second important aspect for object-based change detection is the change detection technique itself. As already mentioned before, we focused on the transformation of the object features space by using the IR-MAD [2]. When applying this transformation to feature spaces with only small correlation between the dimensions, normally no numerical issues can be seen. However, in datasets with high correlations between the dimensions, e.g. in hyperspectral data or an object-based processing using many features, the algorithm results in near-singular covariance matrices which cannot be inverted in a straightforward way. As one possible solution to this problem, it was proposed in [2] to reduce the dimensionality of the dataset and hence to decrease the correlation between the dimensions. Therefore, we implemented a principal component analysis (PCA) transformation of the object's feature space before applying the IR-MAD method.

In the following, we firstly describe the modified multiresolution Segmentation method and introduce the adaptation steps needed to be well integrated into the process of object-based change detection workflow. Secondly, we present the idea of the IR-MAD and show which modifications are necessary to yield a stable behaviour of the algorithm. Thirdly, we demonstrate the performance of the proposed procedures using simulated and real remote sensing data. Finally we discuss the results and give an outlook on further developments.

2. Methodology

2.1. Segmentation adapted for object-based change detection

Earlier studies [4, 5] turned out that segmentation is the crucial step in object-based change detection. A well-known method for this purpose is the multiresolution segmentation algorithm [1], which is available in the eCognition software for object-based image analysis [11]. This technique starts with a segmentation in which every pixel represents a segment. It then uses homogeneity criteria based on color and shape, and a scale parameter in combination with local and global optimisation techniques to iteratively merge neighbouring segments to obtain a segmentation of the imagery [1]. However, object-based change detection requires a segmentation algorithm that, unlike the MRS, similarly extracts objects that have not changed their shape and size between the two acquisition times. The general idea of our work is to create segmentations of the two images I_1 and I_2, acquired at different times over the same area, that only differ in image regions where actual changes took place. Therefore, the MRS is used to generate a segmentation of I_1 that is also applied to I_2 and tested for its consistency. If an inconsistent segment is found, it will be split up. In the following, we will describe how this algorithm was adapted to the problem of segmenting two images of the same area acquired at different times. We propose the following approach:

1. Segment image I_1 using the MRS algorithm.
2. Apply this segmentation to image I_2 and recalculate the heterogeneity of each segment based on the data of I_2.
3. Check every merge, i.e. every segment that consists of more than one pixel, for consistency with the data of I_2 by applying a test criterion. Not only the top-level segments, i.e. the nodes without parents, need to be examined but all nodes in each segment tree except for the leaf nodes.
4. Remove all inconsistent segments using a segment removal strategy.
5. Use the so obtained segmentation as a basis for the MRS algorithm to obtain a final segmentation of I_2.

These steps present a general process which has to be specified in two aspects. First, how can segments of image I_1 be checked for consistency with image I_2? Second, how can segments be removed that are inconsistent with the data of image I_2?

With regard to the consistency tests, we propose three different criteria. The first one, named *threshold test*, examines whether a given segment's S heterogeneity $h(S)$ is within a limit T_{check}. Otherwise, the segment S is marked as inconsistent. The threshold test is the weakest test with respect to changes between the two images. The second test, called *local best fitting test*, tries to repeat the merge procedure with the data of image I_2. Given an exemplary segment structure with segment S_3 having been created by merging arbitrary segments S_1 and S_2, the test assumes S_1 to be a seed and searches for locally best fitting neighbours from the list of merge candidates that has been stored during the segmentation of image I_1. If the best fitting neighbour is S_2, the test is passed, otherwise it is failed. Besides, S_3's heterogeneity has to be within the limit T_{check} as well. Finally, the third test is named *local mutual best fitting test*. It starts like the local best fitting test. But additionally it checks if S_1 is the best fitting neighbour for S_2. Compared to the local best fitting test, this test is even more sensitive. In general, splitting-up segments could be avoided by increasing threshold T_{check}. Then, not all changes between S_1 and S_2 may result in changes of the segmentation.

After testing all given segments for consistency with the image I_2, those segments that did not pass the test have to be handled. We therefore introduce three strategies to remove these segments. The first strategy is named *universal segment removal strategy*. It searches for the top-level segment of an

inconsistent segment and splits it into its elements. As a result, only pixel segments will remain. Obviously, this strategy affects the segmentation intensively.

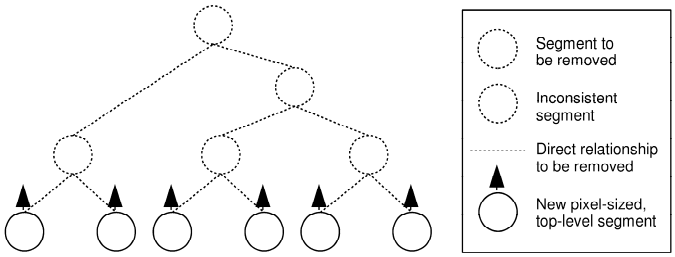

Fig. 1: Universal segment removal strategy.

The second strategy for removing inconsistent segments is the *global segment removal strategy*. Its basic principle is to remove the inconsistent segment and all its ancestors from the segment tree. During this process all remaining segments are considered to be new top-level segments. In this way, the impact on the segment tree is reduced. However, this strategy is very adaptive in creating changes only in parts of the segment tree in which changes can be detected and leaves the rest as it is.

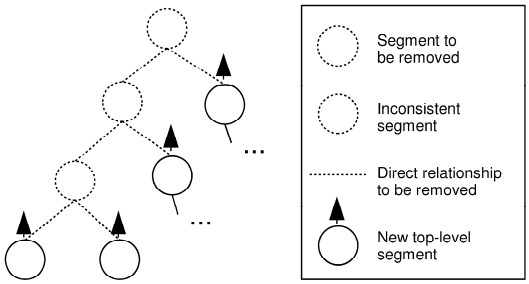

Fig. 2: Global segment removal strategy.

The third and most complex segment removal algorithm is called *local segment removal strategy*. It is developed due to the fact that the global segment removal strategy affects parts of the segment tree which do not necessarily change between different acquisition times. Therefore, we propose an additional method for removing inconsistent segments: Assume *I* to be an inconsistent node. Then remove *I* and its parent *P* from the segment tree. Set *I*'s children C_1 and C_2 as top-level segments and put *I*'s sibling *S* as child of *I*'s grandparent *G*. It has turned out that the local segment removal strategy cannot be applied directly in all possible constellations. This occurs in situations where segment *S* has no common border with segment *U.* See [3] for a more detailed discussion of this issue.

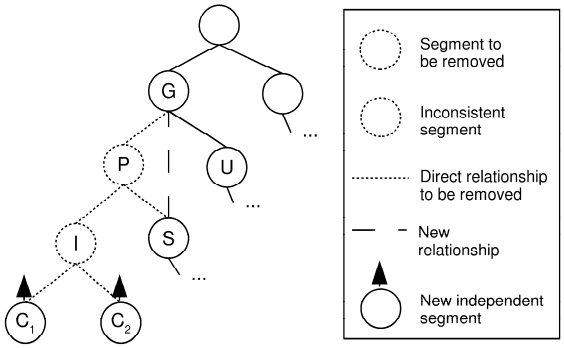

Fig. 3: Local segment removal strategy.

2.2. IR-MAD adapted for object-based change detection

The IR-MAD method is a linear transformation of the feature space aimed at enhancing the change information in the difference image. It models an object's feature vector as random vectors *F* and *G* with *F* representing the information from the first and *G* from the second image. It then calculates the transformation vectors *a* and *b* as the solution of a generalised eigenvalue problem to create difference images M_i as

$$M_i = U_i - V_i = a_i^T F - b_i^T G. \tag{1}$$

with maximum variance under the constraints $Var(U)=Var(V)=1$ and $Cov(U_i,U_j)=0$. The transformed imagery emphasises the differences between the two acquisition times. Moreover, the M_i's, referred to as MAD components, are mutually uncorrelated which has the effect that different components show different types of changes. The sum of squares of standardised variates aproximately follows a χ^2-distribution. Thus, the change-probability can be derived for each pixel or object. For a more comprehensive explanation of the IR-MAD method see [2, 7].

To solve the eigenvalue problem mentioned above, covariance matrices have to be estimated from the imagery, they may not always be invertible [2]. However, the procedure requires this inversion. Therefore, we propose to reduce the dimensionality of the data using the principal component analysis (PCA). PCA is a linear transformation like the IR-MAD method. The difference to IR-MAD is that PCA maximises the variance of a single dataset. Thus, it produces mutually uncorrelated features $U_1,...,U_N$. Hence, only those components containing a significant high variance are used as input for IR-MAD, the remaining features are ignored. In practice, the imagery is transformed and the features $U_1,...,U_M$ are selected if they describe at least 95% of the total variance, i.e.

$$\sum_{i=1}^{M} Var(U_i) \Big/ \sum_{i=1}^{N} Var(U_i) \geq 0.95. \tag{2}$$

For more information on PCA see [7].

2.3. Object-based Change Detection Workflow

Using the segmentation as described in Section 2.1, it is possible to retrieve a segmentation of the imagery with at least three advantages: Firstly, we are now able to integrate shape features into the change analysis. Secondly, the presented segmentation algorithm is robust as it only leads to a different segmentation of image I_2 in areas where using the segmentation of I_1 would not be consistent with the data of I_2. Thirdly, the segmentation results have a high quality because it is not necessary to produce a single segmentation taking both images I_1 and I_2 into account. However, we still receive separate object layers for either acquisition time, which have to be connected in order to obtain a correspondence between the image objects. Corresponding objects are required for applying the IR-MAD transformation, since the IR-MAD algorithm models the objects from I_1 and I_2 as realisation of random feature vectors *F* and *G* respectively. Hence, we need to estimate the parameters using corresponding realisations of *F* and *G*. For that reason we will propose two procedures on how to establish a one-to-one relationship between the segmentations of image I_1 and I_2.
The first procedure, named *directed change detection (DCD)*, associates each segment S_i in I_1 with all segments $T_1,...,T_n$ in I_2 that are overlapping S_1. Since this would not establish a one-to-one relationship, we set the realisations of *X* and *Y* to

$$\begin{aligned} x_i &= f_x(S_i), \\ y_i &= \frac{1}{n}\sum_{k=1}^{n} f_y(T_k), \end{aligned} \tag{10}$$

where f_x and f_y are functions returning the feature vectors of a given segment in the image I_1 and I_2 respectively. Thus, a pair of values (x_i,y_i) is available for every segment S_i in image I_1. This result can now be used to estimate the probability distribution's parameters. An example for a specific object constellation illustrating the method is given in Fig. 4a.
The second procedure that we propose for establishing an object-to-object relationship between the segmentations of image I_1 and I_2 is called *change detection using intersected objects (CDIO)*. The

main idea of this method is to construct a third segmentation by intersecting segments from the segmentations of I_1 and I_2. Given the segments S_1 from the segmentation of I_1 and S_2 from the segmentation of I_2, a segment $S_{1,2}$ is constructed by

$$S_{1,2} = S_1 \cap S_2 \tag{11}$$

This automatically involves a unique correspondence of $S_{1,2}$ in the images I_1 and I_2. Hence, the realisations of X and Y can be calculated straightforwardly for each segment $S_{1,2}$ by

$$\begin{aligned} x_i &= f_x(S_1), \\ y_i &= f_y(S_2). \end{aligned} \tag{12}$$

An example for the application of the CDIO *method* is given in Fig. 4b.

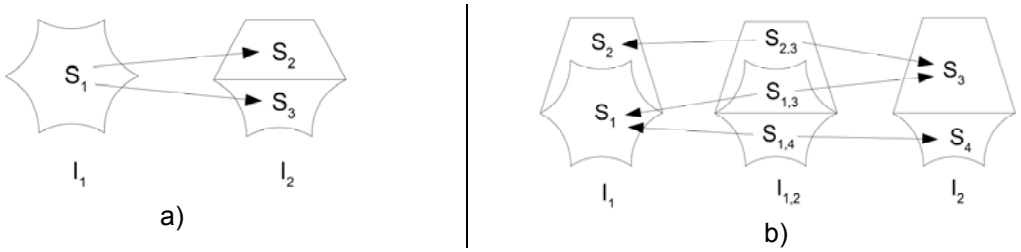

Fig. 4: Object correspondence: a) directed change detection (DCD), b) change detection using intersected objects (CDIO).

For more details on the integration of several segmentations into the change detection process, see [12].

3. Experiments

After having implemented the proposed methods, a test with simulated data was carried out. For the test we used an aerial photography and added artificial changes. In particular a house was copied and pasted to another position in the image. Furthermore some Gaussian noise was added. For reasons of visibility, we will only show a part of the scene in this section. The imagery was segmented using the threshold test and the universal segment removal strategy. To integrate the segmentations into the change detection workflow, the method CDIO was used. Thereafter, a dimensionality reduction using PCA was carried out. This was followed by an IR-MAD transformation and the final change classification using a threshold on the change intensity. Figure 5 shows that our methodology works very well on simulated data as only the relevant changes are detected.

Fig. 5: Result of the change analysis using the method of *correspondence via intersection*; from upper left to lower right: Segmentation of first image, segmentation of second image, change intensity obtained by MAD, change classification using a threshold.

Finally, we tried to apply our methodology to real satellite imagery. Therefore, we used two images acquired in June 2005 and July 2006 over the Olkiluoto site in Finland, with two reactors in operation and the third under construction. As these images were taken almost at the same time of the year, it was expected to reduce influences of different shadows and changes in vegetation which normally cause false alarms. After co-registering the imagery, we applied radiometric normalisation and segmentation using the methodology introduced before with the universal segment removal and the threshold test. This was followed by establishing the object links using CDIO. Using this segmentation as a basis, we carried out a dimensionality reduction using PCA using mean, standard deviation, shape index and compactness as input object features. After that, the IR-MAD was applied on the principal components. Finally, we classified the relevant changes using a threshold on the χ^2-value. The result of this process is displayed in Figure 6. It can be seen that still some false alarms occur in the resulting change map. However, the most relevant changes in the imagery have been detected using the proposed method.

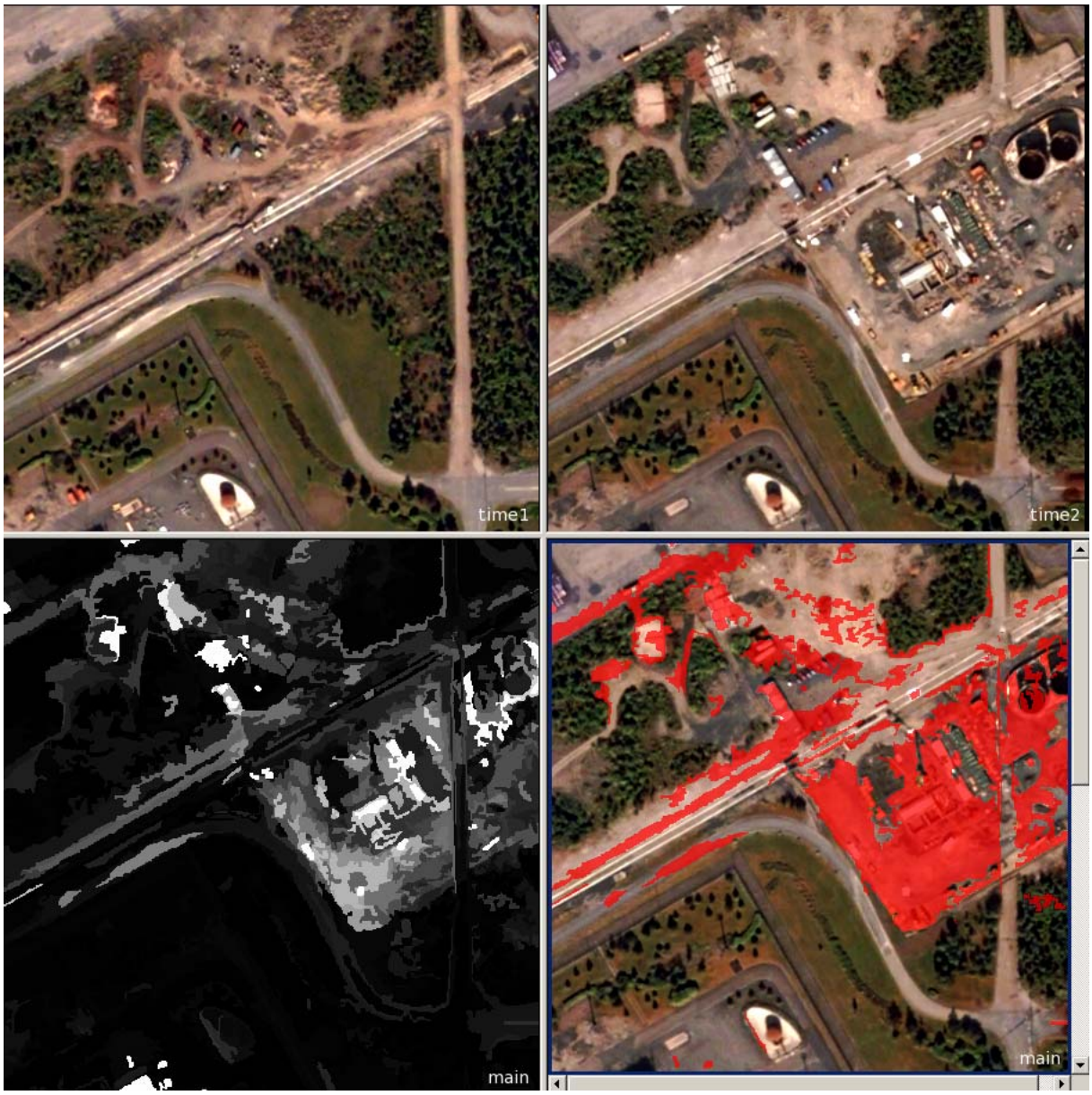

Fig. 6: Change analysis of the Olkiluoto site, from upper-left to lower-right: image acquired in June 2005, image acquired in July 2006, χ^2-values, change classification.

5. Conclusions

We presented some new ideas for object-based change detection for remote sensing imagery. An enhanced procedure for segmentation was presented and implemented into the change detection workflow. Moreover, numerically issues in the IR-MAD method were addressed. These methods showed good results for simulated data. On real satellite imagery, some further tuning of the method has to be done.

Nevertheless, further developments are needed such as new consistency tests and segment removal strategies. Moreover, methods for enabling the user to easily select the parameters, e.g. by using training samples, would be helpful. Finally, the adapted Multiresolution Segmentation has to be implemented as a plugin for the eCognition software so that it can be used in the proposed change detection workflow.

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FACILITY SAFEGUARDABILITY ANALYSIS AND SAFEGUARDS BY DESIGN

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33RD ESARDA ANNUAL MEETING
Budapest, Hungary • May 18, 2011

Why Facility Safeguardability Analysis (FSA)?

SAFEGUARDS-BY-DESIGN OBJECTIVES

1 Design new civil nuclear facilities that meet national and international nuclear safeguards requirements;

2 Make implementation of safeguards at such facilities more effective and efficient;

3 Avoid costly and time consuming redesign and retrofit of facilities;

4 Design facilities so that the misuse of the facility and/or diversion of nuclear material is easier to detect.

DESIGNER'S GOAL

* Reduce the overall project risk (cost, schedule, licensing) associated with International Atomic Energy Agency (IAEA) safeguards.

1

Design Team Challenge

- "Safeguardability" reflects degree of ease with which safeguards can be implemented effectively and efficiently at a nuclear facility
- Need in SBD to link IAEA safeguards requirements with designer's "safeguards best practices" and design options for implementing those requirements
- The challenge in developing the FSA is how to translate this general concept of "safeguardability" into guidance for
  - 1 identifying potential safeguards issues early in the design process;
  - 2 selecting facility-specific SBD best practices and lessons-learned to resolve the safeguards issues in manner likely to meet the requirements of the IAEA;
  - 3 helping the designer anticipate where innovations in his facility design might pose new safeguards challenges that complicate or are unaddressed by standard safeguards approaches; and
  - 4 determining whether facility design modifications or enhancements to specific elements of the safeguards approach offer a more cost-effective solution

2

FSA Evaluation Framework

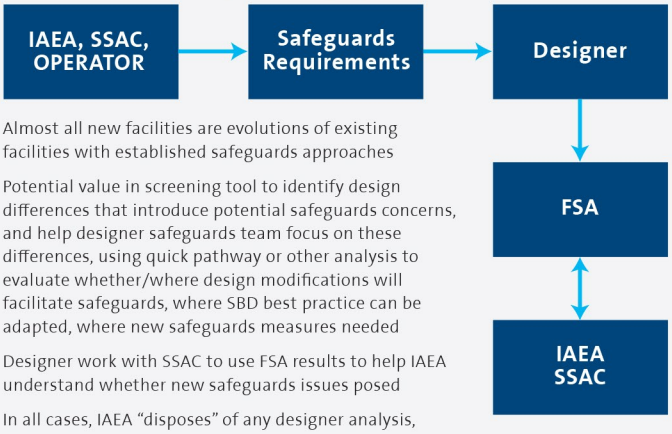
3

Scalable, Targeted Approach to FSA

- FSA for well-understood design, with established safeguards approach and IAEA diversion path analyses—are there places where pathway can be designed away, or is there more effective and efficient ways to safeguard it?
- FSA for innovative elements of new facility design—what are safeguards implications of the design changes, potential for un-reviewed safeguards issue?
- FSA for facilities producing direct use materials—more extensive, inherently complex, but still draw on precedents, best practices
- FSA for first-of-a-kind facility—closest to starting with "blank sheet," potentially greater scope to propose innovative safeguards approaches

4

FSA in Designer Engagement with IAEA



- Almost all new facilities are evolutions of existing facilities with established safeguards approaches
- Potential value in screening tool to identify design differences that introduce potential safeguards concerns, and help designer safeguards team focus on these differences, using quick pathway or other analysis to evaluate whether/where design modifications will facilitate safeguards, where SBD best practice can be adapted, where new safeguards measures needed
- Designer work with SSAC to use FSA results to help IAEA understand whether new safeguards issues posed
- In all cases, IAEA "disposes" of any designer analysis, proposals

5

Next Steps

- The real challenge is moving from this general discussion to an approach that a team of safeguards experts, working as part of the facility design team, can use to do a safeguardability analysis and achieve the benefits proponents of safeguards-by design assert will be realized by both designer and the IAEA
  - How would the designer's team analyze the design, identify pathways, and quickly target new safeguards issues? How and where to make use of documented "best practices" and determine how easily IAEA's safeguards requirements can be met?
  - How can they robustly demonstrate results of analysis?
  - Can this type of analysis be independently verified/validated?
  - What will IAEA look for in FSA analysis if it were presented with such?
- Need a "Test Case" using a well-understood design to demonstrate the process

6

Acknowledgement: This work has been sponsored by the United States Department of Energy, Office of Nonproliferation and International Security (NA-24), Next Generation Safeguards Initiative (NGSI).

PNNL-SA-79599

Characterization of the imaging performance of a portable, coded-aperture, gamma-ray imager for use in enrichment plants

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Abstract:

The imaging performance of a mechanically cooled, portable, coded-aperture, gamma-ray imager designed for international safeguards inspections at uranium enrichment plants has been characterized. The imaging performance of the instrument was quantified by calculating the signal-to-noise ratio as a function of the source distance, position, enrichment, and acquisition time. Based on the expected international safeguards application, importance has been given to the use of uranium standards enriched to less than five percent at source-detector distances greater than two and one-half meters. The implications of the results for the utility of the instrument for safeguards work are discussed.

Keywords: characterization; coded-aperture; gamma-ray imaging; safeguards

1. Introduction

A technical, collaborative effort was established in 2008 to investigate the use of a three-dimensional (3D) laser imaging system combined with gamma-ray imaging for international safeguards applications.[1] The objective of the combined system is to provide the international safeguards community with a tool to verify that the process system design is consistent with the operator's declaration as described in the Design Information Questionnaire (DIQ) and that the nuclear materials are located in the process systems and components in accordance with the operator's declaration. The combined system also has the potential for detecting nuclear materials in areas that are undeclared if the operator has not taken measures to effectively shield the piping/containers where the material is being withdrawn. The possibility of combining different measurement techniques into one tool will optimize the inspection effort and increase safeguards effectiveness.

The work is being conducted in the framework of technical cooperation agreements between the U.S. Department of Energy (DOE) and the European Atomic Energy Commission (EURATOM) and between DOE and the Brazilian-Argentine Agency for Accounting and Control of Nuclear Materials (ABACC). The complete collaboration includes the following DOE national laboratories: Oak Ridge National Laboratory (ORNL), Lawrence Berkeley National Laboratory (LBNL), and Lawrence Livermore National Laboratory (LLNL). The effort also counts on the technical support and expertise from two regional safeguards organizations: EURATOM's Joint Research Centre at Ispra, Italy (JRC-Ispra) and ABACC.

The submission and verification of nuclear facility design information usually occurs during the earliest stages of construction, and the information is periodically re-verified over the operating life of the facility. The design information is verified during construction to define and include the nuclear material processing areas. Regional and international safeguards inspectors continue to re-verify the design information during what are called design information verification (DIV) activities conducted over the life of the plant from construction through commissioning, operation, and shutdown to

decommissioning. The 3D laser component of the system under investigation would be the key instrument to establish the baseline of the design of facilities as well as to detect changes to the 3D volume if shielding is added after baseline measurements have been performed.

Currently, the standard for performing nondestructive assay measurements is to use scintillator or semiconductor gamma-ray detectors to look for the gamma signatures given off by uranium isotopes. Several limitations are encountered with this practice: (1) uranium deposits are sometimes located behind heavy processing equipment, hindering physical access to the source of radiation; (2) an adequate survey of a radiation area requires considerable manpower and time; (3) radiation detectors are omnidirectional in that they do not provide information related to the direction of incident radiation; and (4) when collimation is used to restrict the detector's field-of-view (FOV), systematic errors from uncertainties of alignment and deposit geometries can lead to significant uncertainties in the amount of fissile material present.

In previous studies, the performance of pinhole, coded-aperture, and Compton gamma-ray imaging systems was investigated.[2] The results of several measurements conducted at ORNL did not favor a pinhole imaging system, but they showed that coded-aperture imaging was very promising for locating sources of enriched uranium. Other work showed that Compton imagers were promising for locating sources with higher energy emissions, and work by L. Mihailescu et al.[3] has demonstrated the feasibility of combining both types of imaging at intermediate energies.

Tests in an operational facility at ORNL were conducted using both the 3D laser scanner and the portable, coded-aperture gamma-ray imager.[4] The principle of combining outputs from two different commercially available technologies has been proven and demonstrated. The coded-aperture HPGe detector was capable of detecting a line source of 7.54 g of ^{235}U, 4.46% enriched, at a distance of 1.8 m during 200 seconds of counting time. Some of these results were obtained with a large laboratory prototype imager[5] that has recently been upgraded to a highly portable, mechanically cooled instrument of comparable detector area.[6]

The final task of the project aims at conducting measurements in real facilities. In preparation for these field trials, the new HPGe-based, coded-aperture imager is currently being characterized to measure lower energy sources at distances greater than 2.5 m. This paper presents the results of those measurements.

2. Instruments Used in this Project

A 3D laser scanning system developed by JRC-Ispra for DIV is being used to create 3D maps of rooms and objects and identify changes in positions and modifications with a precision on the order of millimeters (see Figure 1).

The laser scanner provides a rapid and accurate locus of points off of all objects, walls, ceilings, floors, etc. within its FOV. This information can be used to compare the location and configuration of process equipment to plant design documents and/or prior measurements, providing inspectors a direct measure that unauthorized modifications have not been made to the equipment. The laser scanner data can also be combined with the images from the gamma-ray imaging instruments to provide the likely location (particular piping, equipment, etc.) of radioactive materials (see Figure 2). The presence of such materials in unexpected locations is a strong indication of illicit plant operations.

Figure 1: A Zoller+Fröhlich Model 5006*i* 3D laser that is being used to complete the project. The battery is part of the single unit, which also stores the image. The unit can be transported in two containers: one for the laser head and another for the tripod.[7]

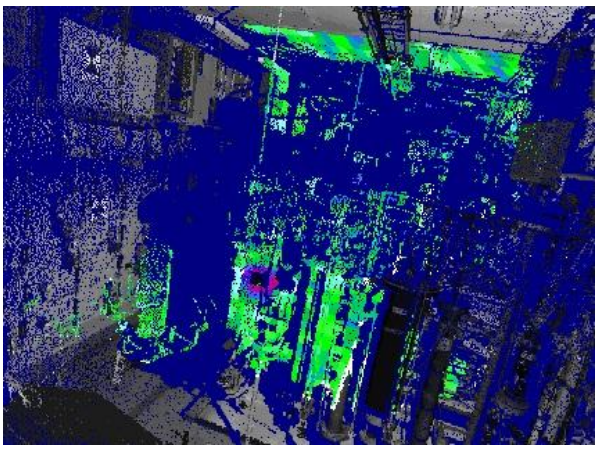

Figure 2: Laser-scanner data combined with a gamma image.[6]

The coded-aperture, gamma-ray imager is based on a commercially available, mechanically cooled HPGe double-sided strip detector. It provides high-resolution spectral information specific to individual material deposits (see Figure 3) allowing application of classic non destructive assay (NDA) techniques to determine deposit isotopics. The relatively mature coded-aperture imaging technique[8] uses a shadow mask that is 50% open area to encode the scene (radiation image) onto the position-sensitive, planar HPGe detector. The instrument has a 5-X zoom capability to allow flexibility in obtaining images remotely (standoff distances from 1 to 10 m from the process equipment). It is mounted on a tripod and operated from a touchscreen interface, providing 2D false-color, gamma-ray images that can be manipulated for spectral information in real time. The instrument includes a visible-light stereo camera aligned with the gamma-ray imager field-of-view (see Figures 4–5). It provides 2D black and white, visible-light images and 3D point data similar to, but lower resolution than, those available from the laser scanner. The black and white image is displayed with a rectangle indicating the gamma-ray imager, zoom-specific field-of-view to aid with pointing the instrument. The 3D locus of points is used to overlay the gamma-ray and laser scanner data. The coded-aperture imager has been optimized for use at lower energies (200 keV and below).

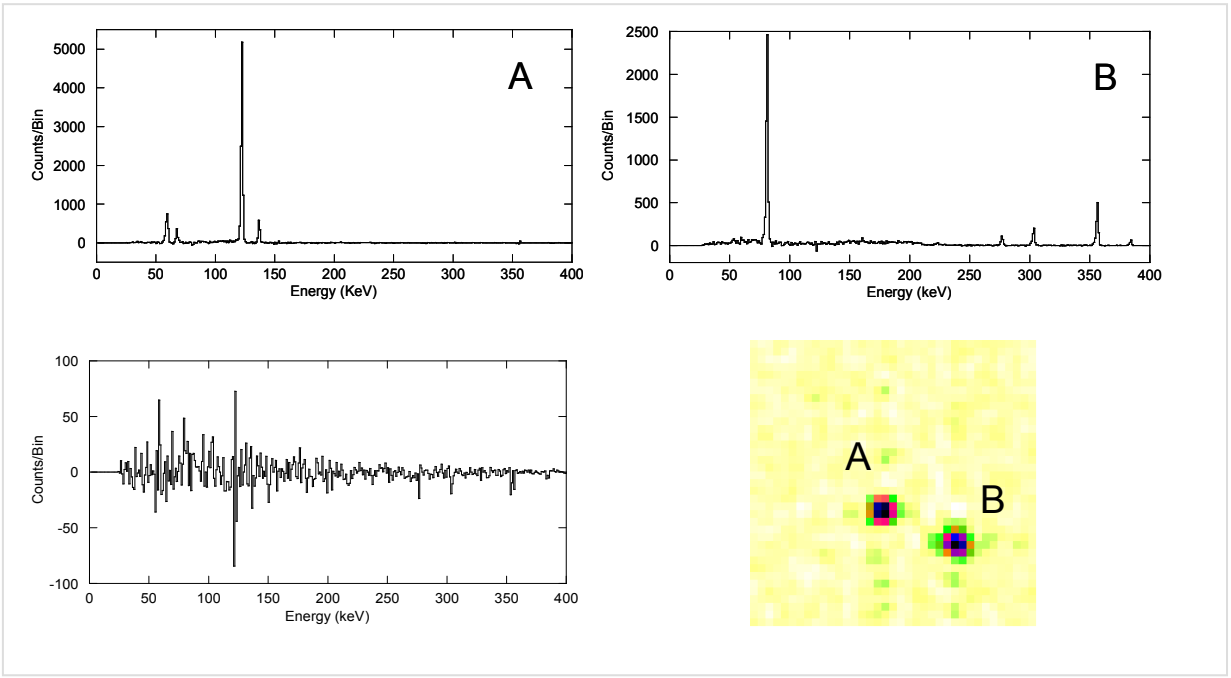

Figure 3: Gamma-ray image of a ^{57}Co (A) and ^{133}Ba (B) point source (bottom right) and the spectra obtained from the central pixels (as labeled) of the sources and an arbitrary pixel (bottom left).[6]

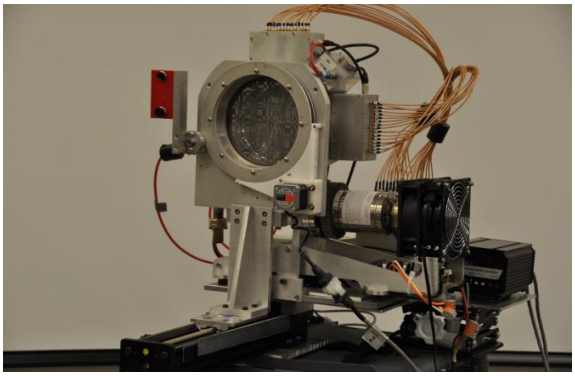

Figure 4: Mechanically cooled HPGe detector with coded-aperture mask and visible-light stereo imaging system (red box beside the mask) with the cover removed.[7]

Figure 5: The instrument including cover and touch screen embedded computer (left side of box).

3. Results of Tests Conducted with the Coded-Aperture Imaging System and the 3D Laser Scanner

A one-week measurement campaign was conducted in a chemical makeup area located immediately above hot cells in which neutron-activated targets from ORNL's High Flux Isotope Reactor are dissolved for extraction of the activation products. A complex of piping and valves allows operators to produce aqueous solutions used for target dissolution and product extraction in the hot cells. Visible images of the complex plumbing in this building were generated using the Zoller+Fröhlich Model 5006*i* 3D laser imager acquired by ORNL. Sealed sources of uranium enriched in the ^{235}U isotope were placed within the framework of the plumbing to simulate pipes or valves containing nuclear materials for imaging.

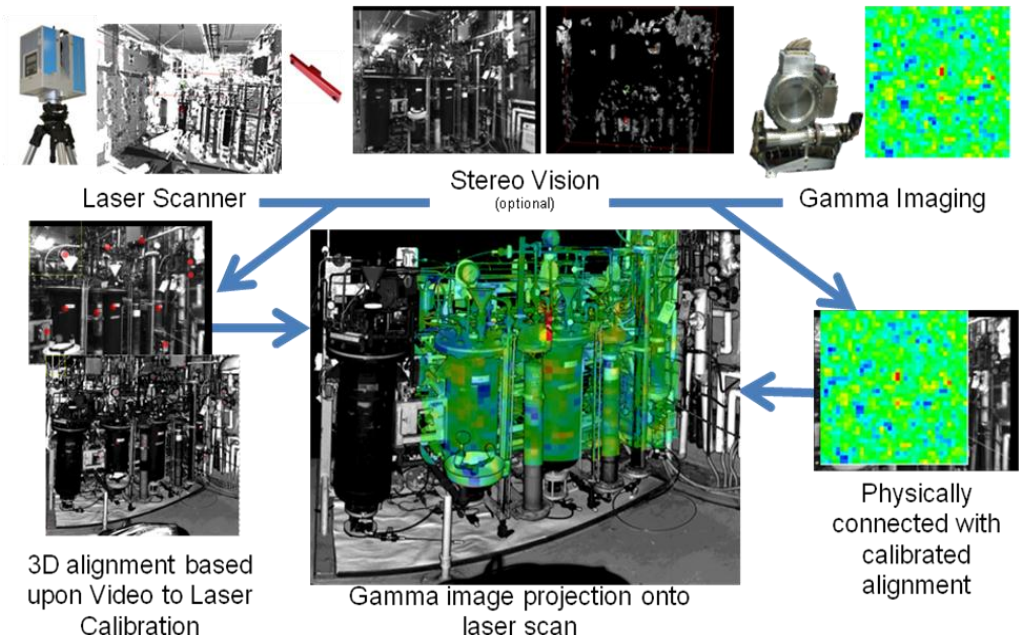

Figure 6: Process of how gamma image is combined onto the 3D laser scan.[7]

The sequence of images in Figure 6 shows how the gamma image is combined onto the laser scan. In this sequence, the source was located at 1.8 m distance from the detector. It was 7.54 g ^{235}U enriched at 4.46%. The gamma-image sensor and stereo-image device are physically connected and calibrated to one another so that the two images can be overlaid. The stereo-vision rig serves as a bridge between the gamma camera and the laser scanner. It is physically aligned and calibrated to the gamma-imaging device. Common alignment allows the gamma data to be projected onto the 3D laser scan.

4. Characterization of Detector

To characterize the detector, its response was found by calculating the signal-to-noise ratio (SNR) for a low-enriched uranium (LEU) standard. The SNR was calculated by dividing the number of counts in the image pixels in which the source was located by the root mean square (RMS) of the counts in the image pixels in which the source was absent. This is a first order approximation derived from the imaging properties of coded apertures that means each of the pixels in the image has approximately the same variance. This is a direct result of the cross-correlation function used with the measured shadow pattern and the known mask function:

$$I_{i,j} = \sum_{m,n} M_{i+m,j+n} D_{m,n} \qquad (1)$$

Here $I_{i,j}$ is the number of counts in pixel (i,j) of the image, and $M_{m,n}$ and $D_{m,n}$ are the mask function and detector counts, respectively. The mask function is either plus or minus one depending on whether the mask pixel is either open or closed, respectively. As can be seen, all counts in the detector contribute to every image location.

It is also important to note that the SNR values will depend on both the radiation from the source as well as the background radiation within the imaging environment. Another method of calculating the SNR is currently being investigated for comparison.

A uranium standard enriched to 4.46% ^{235}U was used for these measurements. The standard consisted of 200 g U_3O_8 powder with a density of 5.2 g/cm^3 contained in an aluminum can of right cylindrical geometry with a diameter of 70 mm. The bottom of this source cylinder was oriented parallel to the surface of the imager. Self-attenuation effects were not included as part of the analysis. Data were acquired for this source located in the center of the FOV at 3, 4, and 5 meters with a constant focal length (spacing between the mask and detector) of 7.5 cm. The respective pixel sizes for this focal length and source distances of 3, 4, and 5 meters are 5.7 cm, 7.5 cm, and 9.4 cm.

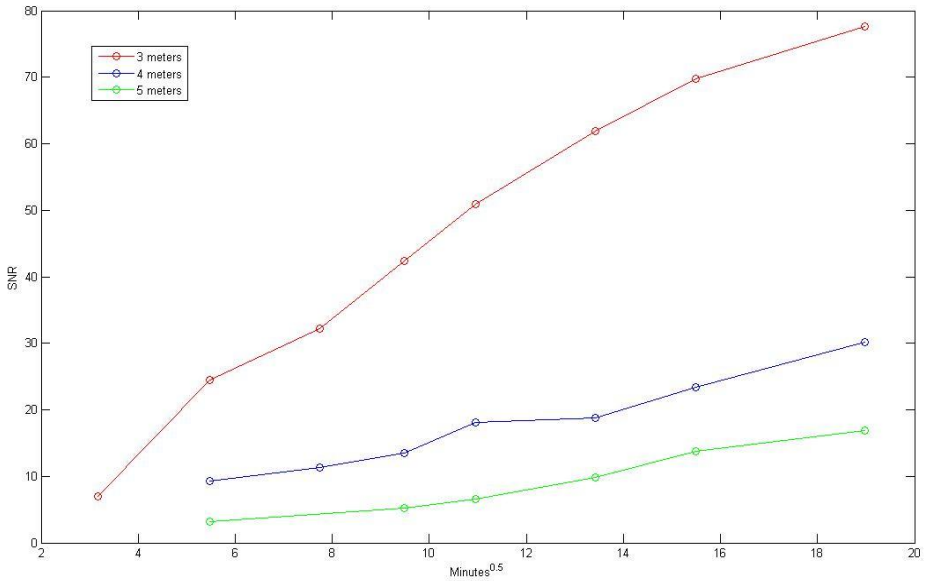

Figure 7: Linear relationship of SNR vs. square root of the acquisition time for a 4.46% enriched uranium standard at 3, 4, and 5 meters.

The data were acquired in list mode so it could be replayed with different parameters. In this case, each long run was subdivided into shorter time intervals at each distance to generate the data in the plot shown in Figure 7. The results are based on an energy window limiting the data to only the 186-keV peak of ^{235}U. The detector data were then exported and analyzed to calculate the SNR in the manner described above. The results show the expected linear relationship between the square root of the acquisition time and the SNR. From the plots, an appropriate acquisition time can be determined to achieve a desired SNR. For this analysis, a SNR value of five was chosen as the minimum needed to signify detection. This SNR corresponds to a false alarm probability of approximately one in 2,000,000. However, each image contains ~ 350 resolution elements so that the true false alarm rate per image is one in 5,500. Once the necessary acquisition time to achieve this value of SNR was determined for each distance, a plot was generated of distance versus time (see Figure 8). The acquisition time on each plot signifies the total time needed for measurement as it includes both the mask and anti-mask data. Due to self-shielding effects, the time for detection of a particular mass of 4.46% enriched uranium depends greatly on the geometrical configuration of the source.

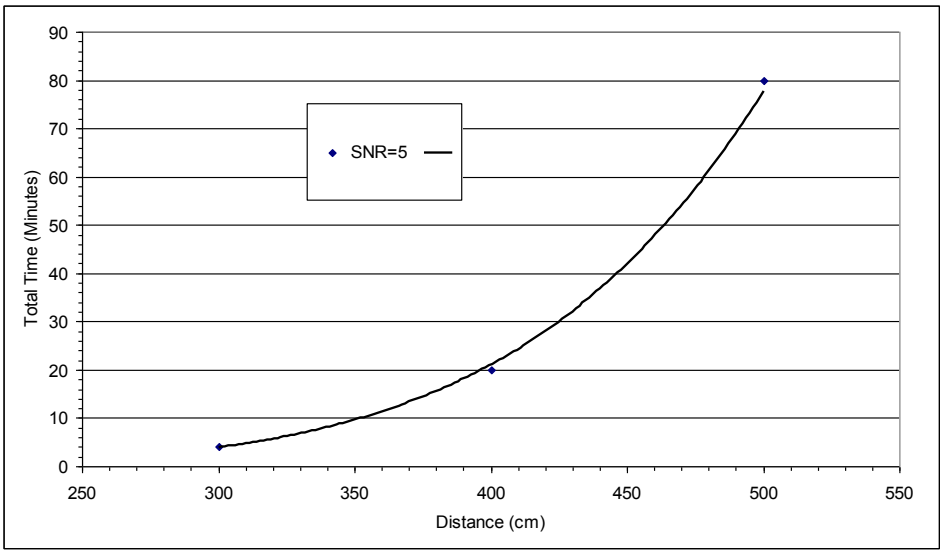

Figure 8: Plot of distance versus time for a SNR ratio of five for a 4.46% enriched uranium standard.

5. Discussion

Additional data are being collected and analyzed to further study the performance as a function of enrichment and other system conditions. For instance, the efficiency for detection of a source will vary as a function of location in the FOV. This has been probed using 97% enriched uranium and is shown in Figure 9. These data were collected at a distance of 3 m with a 7.5 cm focal length. The falloff in sensitivity at the edges of the plot is larger than expected. There are several effects that contribute to this: first, there is a falloff in projected detector area as the source is moved off axis, which varies as the cosine of the angle to the source, and has been corrected for in the figure. Of the remaining falloff, approximately half can be attributed to self-collimation effects due to the finite thickness of the mask. These are estimated using ray-tracing simulations. The remaining reduction in efficiency is still under investigation and is thought to be due to the finite thickness and transparency of the detector reducing the efficiency of detector pixels at the edge of the shadow region.

As already mentioned, for a given measurement, the system performance will depend on the amount of background radiation compared to the amount of flux from the source. This can be influenced by

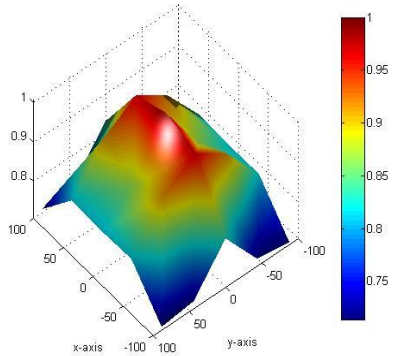

Figure 9: Normalized data of 97% enriched uranium showing the efficiency for detection as a function of source location within the FOV.

selecting different regions of interest in the spectral data. The relatively unshielded material used in these studies shows not only the 186-keV line but also flux at 144 keV and atomic fluorescence lines around 100 keV. The relative proportions of these lines will depend on the enrichment and geometry of the material (see Figure 10)**.** The results in Table 1 show how the selection of different energy windows influences SNR of the results. Here, the SNR is calculated by dividing the counts from the source region of the image by the square root of two and then multiplying by the one-sigma fluctuations observed in the off-source regions of the image that have the same number of pixels as the source region (All of the data were collected at a distance of 3 m, but the amount of material, integration times, and source location within the image were not sufficiently controlled to allow detailed comparison of absolute numbers between the samples.). Of course, the emissions from the process equipment will be preferentially shielded at lower energies, reducing the contributions from the lower energy lines. While collecting data using other than the 186-keV line can reduce the dwell time required to make detection, optimization for a given situation will need to be performed.

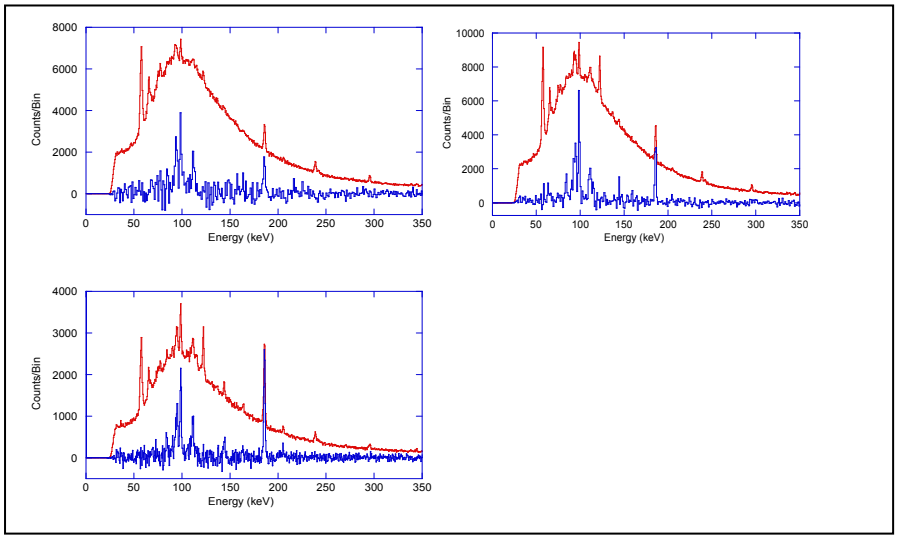

Figure 10: Spectra of U samples with different enrichments. The red (higher) spectra are from the detector as a whole. The blue (lower) spectra are from the source region of the image. Note the difference in relative line intensities for the different samples of 1.94% (top left), 4.46% (top right), and 93.17% (bottom left). The localized spectra are inherently background subtracted due to the imaging process of Equation 1.

Enrichment	All Data	181–189 keV	90–101 keV 181–189 keV	90–101 keV 141–146 keV 181–189 keV	90–189 keV
1.94%	12.8	9.5	16.3	15.9	12.3
4.46%	18.2	16.8	29.5	31.1	21.6
93.17%	28.6	36.3	44.5	42.8	31.1

Table 1: SNR versus enrichment and spectral regions.

6. Conclusions

While the combined data from lidar and gamma-ray images provide an important observable to ensure safeguards compliance, the optimum use of the gamma-ray images requires continued investigation. As with any radiation measurement technique, the sensitivity of the images will depend

on the size of the detector, its distance from the process equipment, and the local background, as well as attenuation by process equipment, deposit enrichment, and geometry.

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Boron-10 lined Tubelet proportional counters for waste assay: a feasibility study

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Abstract:

The global shortage of helium-3 gas presents a serious problem for neutron measurements in the contexts both of safeguards and plant operations. This paper reports upon a collaborative feasibility study which investigates the potential for using proportional counters based on boron-10 lined "Tubelet" clusters as an alternative to helium-3 proportional counters.

Boron lined technology has gained acceptance within the homeland security sector and this paper evaluates the extent to which similar solutions could be developed for the more demanding field of safeguards instrumentation. The primary neutron techniques used for passive non-destructive assay are described and the demands which they place upon detectors, subsystems and instruments are discussed. In this context an experimental programme is defined which enables boron-10 lined Tubelets to be evaluated for safeguards applications.

This work is being undertaken by the TenBee collaboration which receives funding from the UK Technology Strategy Board for R&D to support the civil nuclear industry.

Keywords: boron; helium; detector; assay; neutron

Spent Fuel Assembly Gamma-Neutron Inspection System. SICOM NG FA

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Abstract:

One new hardware and methodology to implement in the radiological inspection of the spent fuel assemblies previous to the dry intermediate storage in Spain is described.

ENUSA has designed in collaboration with TECNATOM the SICOM-NG-FA new equipment for radiological characterization of spent fuel assemblies. The system has been qualified with on-site measurements in Spanish PWR (17x17) NPP and has been analyzed and satisfactory verified in 2010 by the Spanish radioactive waste control organism, ENRESA. The system permits a total inspection of the radiological emissions in the whole fuel assembly length, either for PWR or BWR design. The position of the detectors in front of each face of the assembly is fully controlled by a positioning system and the axial position of the collimators is registered automatically and synchronized with the advance of the collimators. The four faces can be inspected with the gamma and neutron digital spectrometers installed in the system.

The implementation of specific analysis software and codes for each fuel design provides the minimum achievable uncertainty for burnup determination. From the point of view of the safeguards control, this equipment can provide a good discrimination capability to be implemented in production mode. From the point of view of the detection of anomalies in burnup profiles, this equipment is also able to offer a complete characterization.

Designed to be operative and economic during on-site inspections the system does not require N_2 for cooling and is light, compact and easy to install over the spent fuel pool racks. The short inspection time (<30 min for scanning the four faces of the fuel assembly) would permit to inspect a high number of fuel assemblies without much interference with the plant activities.

Keywords: Inspection system; spent fuel; non destructive analysis; digital spectrometer; new hardware

1. Introduction

ENUSA has designed in collaboration with TECNATOM the SICOM-NG-FA, new equipment for radiological characterization of spent fuel assemblies [1].

A goal when designing this equipment for inspection of irradiated fuel assemblies was to obtain a direct measurement able to give the maximum information of each fuel assembly, not only burnup, but also of other aspects that can be obtained from radiation through inspection, as cooling time estimation, determination of fuel assembly end of cycle power or nozzle ^{60}Co radiation level evaluation.

Another requirement for system design was to get an easy installation of the equipment on the racks allowing its stay for some time under water. Operation without N_2 cooling was also established as a condition. The safety of the fuel operation was evaluated in all its aspects, with the collaboration of experts in nuclear fuel operations in NPP's and in fuel design.

Once every aspect of the acquisition system was optimized, the definition of the specific

spectrum analysis software has been completed, the extraction of net counts was adapted to the specific characteristics of the measure, the results did not disappoint. In turn, the nuclear design engineers made the contribution to create accurate algorithms applying to each isotope and fuel design.

The system has been qualified with on-site measurements in Spanish PWR (17x17) NPP [2] and has been analyzed and satisfactory verified in 2010 by the Spanish radioactive waste control organism, ENRESA.

The qualification results were satisfactory and gave an acceptable uncertainty in the burnup measurement compared with nuclear design values.

Therefore SICOM-NG_FA may be considered an appropriate option to apply in the controls on fuel irradiated prior to dry storage.

2. Geometry control

Uncertainty associated with variations in the relative distance between the source and the detectors is reduced to the maximum achievable by introducing positioning in the horizontal plane.

The axial position is also recorded as the fuel assembly moves along the detectors, the speed is also monitored and all this information is synchronized with the recording information of the profiles. In figure 1 a picture of the system is included.

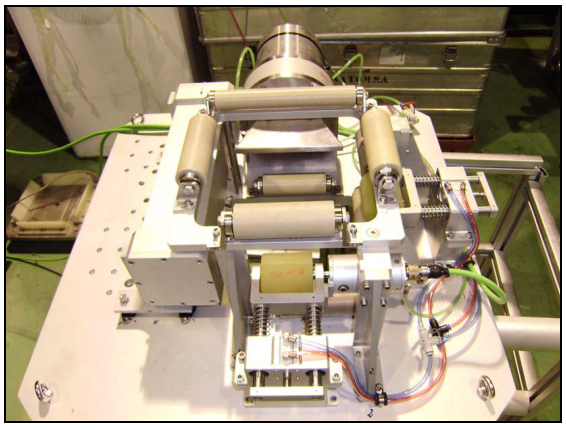

Fig 1: SICOM-NG-FA: Collimators Gamma and Neutron, positioning system and register of axial advance.

Since the collimators have been designed to optimize efficiency and spatial resolution as the profiles are registered, the geometric control is the right tool for the subsequent detection of anomalies in fuel assemblies, but also to reduce the uncertainty in the determination of burnup through the neutron or gamma measurements.

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3. Measurement capabilities: Gamma and Neutron.

Gamma and neutron measurements for characterization of spent fuel can be performed with the SICOM-NG-FA. Both measurements are complementary and provide meaningful information:

Neutron Measurements: A fission chamber detector is implemented in the system. It can provide information on burnup fuel, not only from the peripheral rods but also from the interior rods. This measurement is an excellent indicator of burnup by its dependence with the neutron flux. Spectral recording is performed simultaneously with the acquisition of a profile (cps). Figure 2 shows an example of neutron data registered in one face advance profile.

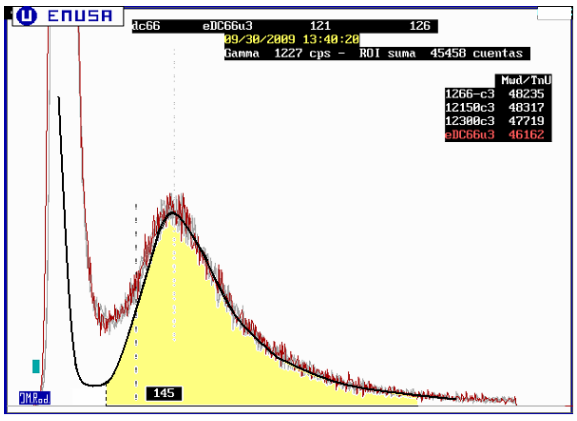

Fig 2: Neutron data (yellow)

Gamma Measurements: A CZT detector is used for gamma measurements. It provides isotope discrimination and net counts of ^{106}Ru, ^{137}Cs, ^{134}Cs and ^{154}Eu. This is not a very deep inspection, radiation comes from the outside parts of the fuel, and this channel gamma provides a high spatial resolution in the gamma total profile measurement. The simultaneous recording of total gamma cps and spectrum of the profile is essential.

Figure 3 illustrates an example of Gamma Spectra registered in one face advance profile (2min)

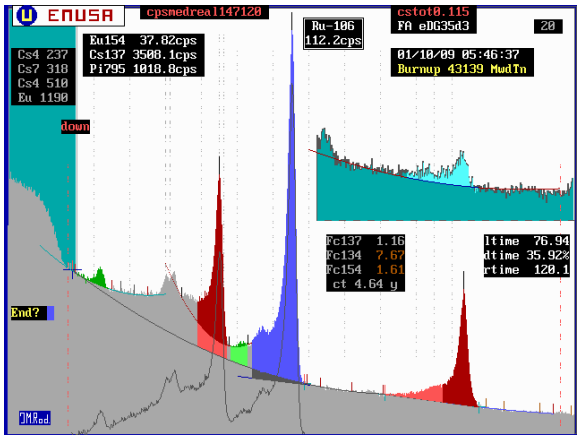

Fig 3: Gamma Spectra registered in one face advance profile

The simultaneous register of spectrum and profile in neutron or gamma measurement channels allows that all the nuclear material of the fuel assembly can be involved in the same grade in burnup determination. Additionally, this methodology also improves quality control records.

4. Processing, Analysis and Data Reduction.

For the processing of the data each measurement channel (gamma or neutron) is managed by digital processors 2+ suitable for measuring high count rates.

The digital processors (figure 4) can be configured by software. This instrument provides the guarantee of stability and registration of the entire spectral band. The acquisition is always made in terms of cps profile & spectrum for each channel gamma or neutron. The quality of the scanning of one fuel assembly can be unambiguously established through the analysis of the recorded data once crossed with data of axial speed and position.

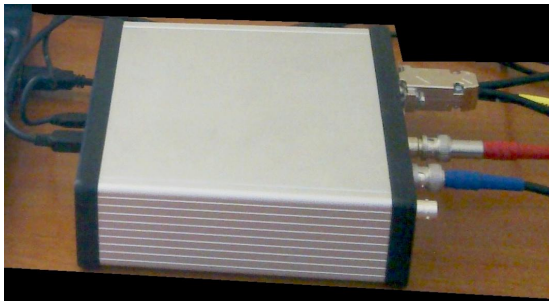

Fig 4: 80MHz ADC, Ethernet Digital Processor

The analysis of neutron data in terms of cps is quite simple thanks to the refinement of the neutron collimator design, but the conversion of counting records to burnup units requires the application of a sophisticated proprietary algorithm for each fuel design. The results thus obtained are usually the best estimation of the burnup media.

In the gamma case the opposite happens but although a good extraction of net counts is not easy to achieve with CZT detectors a method has been specifically designed for this purpose. Moreover, the algorithm that relates measurements with the burnup is not complicated and corrections can be very accurate. Figure 4 shows superposition of “4 profiles/face” for one FA (gamma total, 2 minutes profile)

So when a fuel assembly is measured on all four faces, two burnup determinations per face (gamma and neutron) are generated, therefore average burnup values can be calculated from eight measurements. It is expected that with the results of additional campaigns the average burnup can be calculated with a higher accuracy.

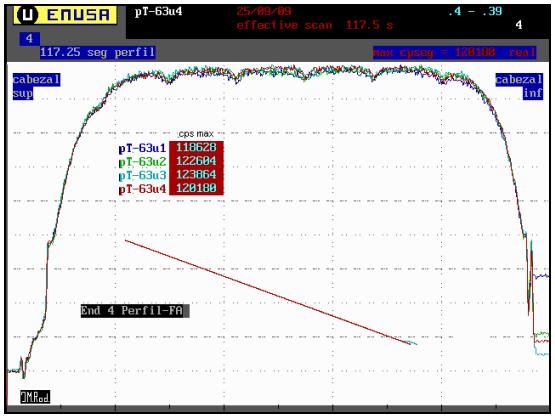

Fig 5: Shows superposition of “4 profiles/face” for one FA (gamma total, 2 minutes profile)

One study has demonstrated the feasibility of the option of setting the final measurement, regardless of cooling time and irradiation history coming from the reactor data; this is achieved from the values of net counts of the isotopes: ^{106}Ru, ^{137}Cs, ^{134}Cs & ^{154}Eu, combining them with the neutron measurement results. Nevertheless, a more extensive number of fuel assemblies measurements will be necessary to properly determine the uncertainty for this unusual choice.

5. Specific features of the system.

The equipment has been designed with the goal of having an additional system for radiological characterization of irradiated fuel with the following distinctive features:

• It combines in the same sequence burnup evaluations and face profile, with gamma and neutron collimators designed for optimum spatial resolution, to easily detect any irregularity.

• It gives robustness and easiness to operate, providing enough safety guarantees in the movement of fuel thanks to control of the measurement geometry in the advance.

•It offers analysis software specifically designed for each detector head, having all the records of the measurements, either gamma or neutron in spectrum mode. Additionally specifically algorithms designed for each fuel assembly type can be available.

• All nuclear material is involved in the average burnup evaluations and expressions of the profiles. All significant types of radioactive particles emerging from the fuel assembly take part in the evaluations.

• Inspection of 4 faces can be performed allowing low uncertainties in the average burnup measurements, as well as availability of the most complete radiological information from the fuel assembly.

• Simultaneous visual inspection to profile measurements could be also performed to comply with increasing demanding in visual characterization prior to dry storage.

6. Conclusion.

The characterization system of ENUSA for irradiated fuel previously to dry storage has been described. Their capabilities make this system suitable for characterization purposes of irradiated fuel and safeguards control.

Although there is not an International Standard for radiological control of irradiated fuel previously to dry storage [3], this system enables the detailed characterization of the fuel and may be a reference to expand the information available about irradiated fuel before storage.

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25 Poster session II

Validation of a new software application for Hybrid K-Edge/XRF Densitometry

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Abstract:

The Hybrid K-Edge Densitometry (HKED) system is designed to determine the uranium and plutonium concentrations in dissolver and product solutions during fuel reprocessing as an alternative to destructive (chemical) analysis. The HKED technique can give approximately the same measurement precision as destructive chemical analysis methods, yet is much simpler and faster to use. The current generation of the Hybrid K-Edge/XRF Densitometry software was developed for use on the VAX/VMS platform. However, advances in computer and software technology suggested the porting of the VMS-based software to a PC platform running under the current versions of the Windows OS. The new software was designed for facility and industry level use, and was therefore subjected to extensive testing and quality assurance review. In addition the analysis was tested using both measured and synthetically generated data that stressed the algorithms. Since the VMS-based version of the software has proven to be very reliable in terms of providing accurate and precise data and thus accepted worldwide by the international safeguards community the algorithms used in the analysis remained unchanged and were imported into the new software in order to ensure traceability of results. A side by side comparison of the analysis was then performed for the two software packages. The validation of the software was based on measured data obtained with a commercially available HKED densitometry instrument. Each of the analysis options that are available in the software was tested, including Hybrid K-Edge Densitometry, Uranium and Plutonium K-Edge Densitometry (UKED and PuKED), and Uranium and Plutonium XRF Analysis (UXRF and PuXRF). During the test a number of spectra were used to calibrate the counting system and perform the analysis. These results were then compared to the same data obtained using the VMS-based software in order to validate that all analysis algorithms had been imported correctly. The comparison results presented in this paper showed excellent agreement between the two versions of the software.

Keywords: Hybrid K-edge densitometry; KED, XRF analysis

1. Introduction

Canberra's Hybrid K-Edge Densitometry (HKED) system is based on a system developed by Ottmar and Eberle at the Institute for Transuranium Elements (ITU) in Karlsruhe, Germany [1, 2] and is designed to determine the uranium and plutonium concentrations in dissolver solutions during fuel reprocessing as an alternative to destructive (chemical) analysis.

The HKED system consists of two sub-systems, which may be used either separately or in combination; they are:

- K-edge Densitometry sub-system (KED) to measure the X-ray transmission discontinuity across K-edge energies

- X-ray Fluorescence (XRF) sub-system to either determine the elemental concentration ratio for HKED or to determine the absolute elemental concentrations for low-concentration solutions

Special Hybrid K-Edge/XRF Densitometry software has been developed in the past to automate the process of measuring and analyzing U/Pu sample solutions. The original HKED software is VMS-based, but with increasing concerns for the obsolescence of this platform, a new PC-platform based software has been developed. The new software has an easy-to-use graphical user interface for all functions, including setup, calibration, sample measurement, and system maintenance. While not directly visible to the user, the algorithms used in the analysis remained unchanged in order to ensure traceability of results across the transition. Also not directly visible is the underlying architecture of the software which allows for the future addition of new analysis algorithms when needed.

2. Hybrid K-Edge Densitometry software

The detailed description of the HKED software features and its evolution from the VMS-based version to Windows-based software is given elsewhere [3]. This paper presents the Windows-based software validation test results and only gives a brief description of the software functionality.

2.1. VMS-based

The Main view of the existing VMS-based HKED software is shown in Figure 1. The software automates the process of measuring and analyzing U/Pu sample solutions by controlling gamma-ray detectors with an X-ray source to measure X-ray transmission at K-edge energies and to perform XRF spectroscopy.

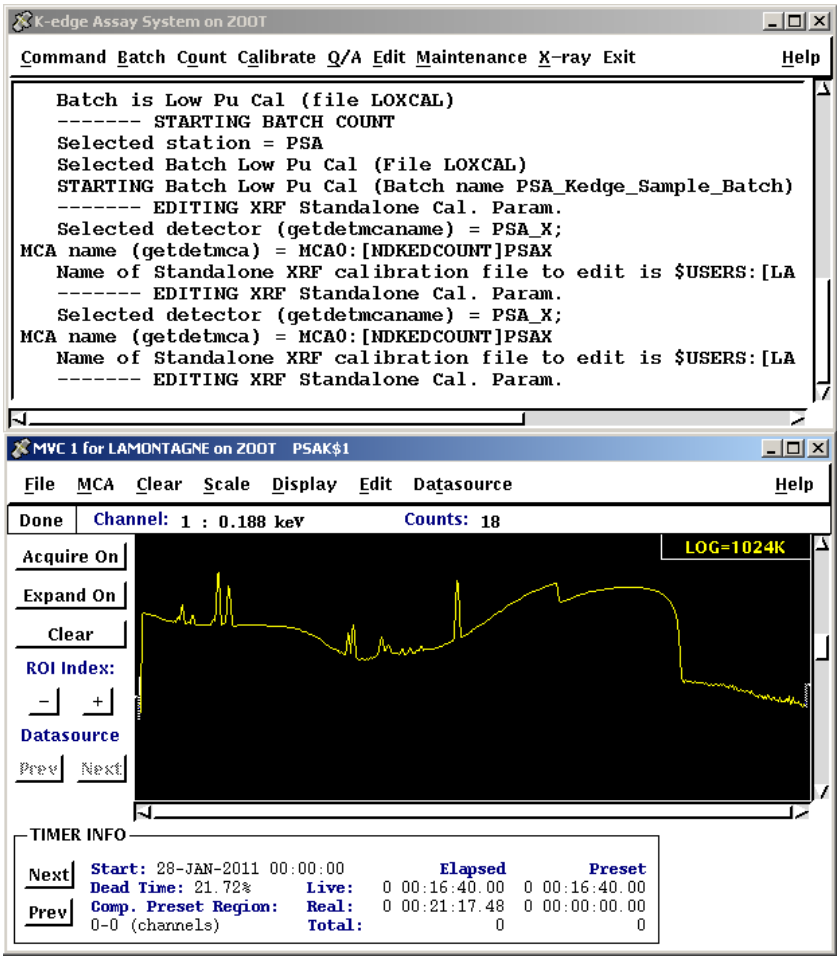

Figure 1: Main view of the VMS-based HKED software

The software allows the control of NDA measurement stations by a computer or workstation running VMS DECWindows Motif and using Ethernet protocol to control the Multi-Channel Analyzer hardware.

Although this software has been widely used and proved to provide very accurate analysis results, the main limitation of the existing VMS-based HKED software is in the operating platform itself. The VMS operating system is two-decade old technology, which today is becoming less familiar to the majority of new users and software developers. In addition, new challenges to the HKED technique itself can be anticipated on the horizon that will likely require changes and additions to the analysis capabilities of the software. Some examples of this are sample solutions containing multiple elements of comparable concentrations (e.g., MOX solutions), the presence of multiple actinides and an increased presence of fission products in the solutions. These challenges have long anticipated the need to transitioning the existing application algorithms to the Windows platform.

2.1. PC Platform-based

The new Windows-based software consists of two main applications:

- Operations application
- Data Review application

The Operations application, which is shown in Figure 2, is used to configure a HKED counter, perform calibrations and regular assays. It can also be used to operate the Sample Changer and to control the X-Ray generator through serial communications. The application provides access to the declaration and certificate editors, sample containers editor, calibration and assay settings. It supports multiple calibrations for different sample types.

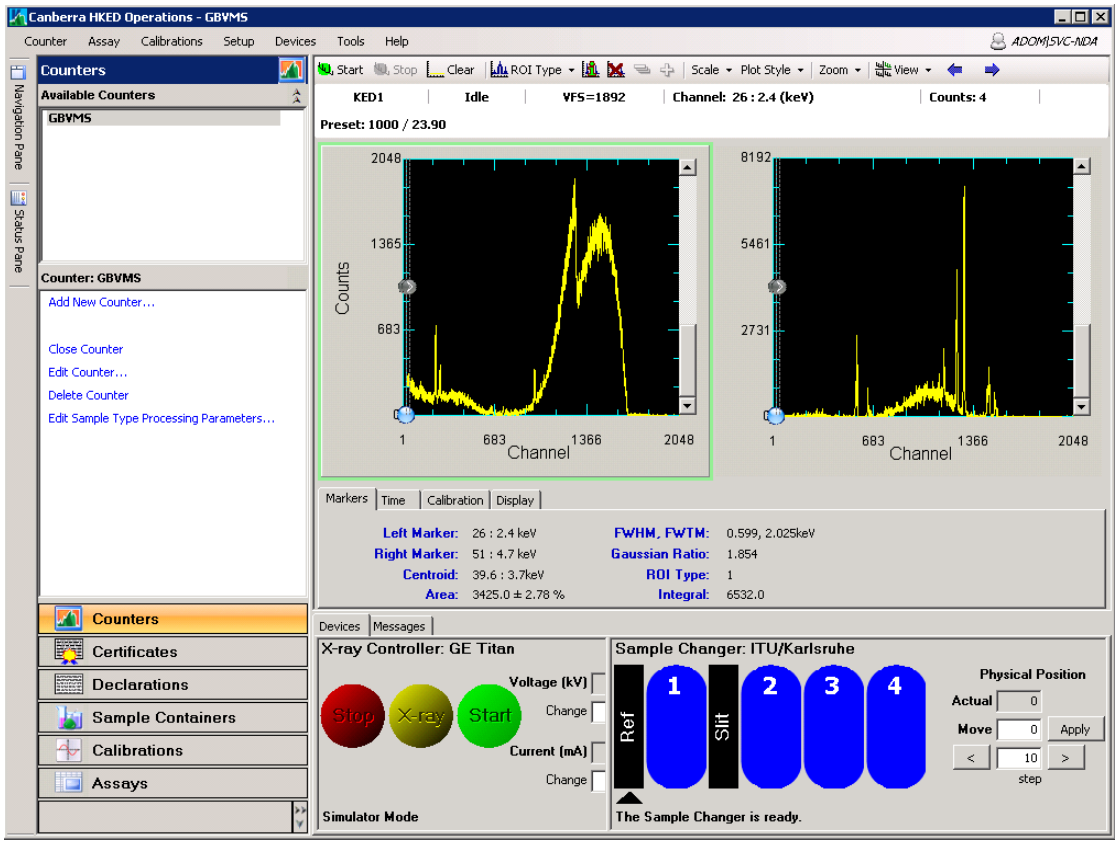

Figure 2: Main view of the Operations application in Windows-based HKED software

The Data Review application is used to review previously acquired counts and gives an option of reanalyzing them if necessary. This application is also used to produce QA plots and reports based on the predefined procedure for the QC check measurements. The main view of the Data Review

application is shown in Figure 3. In this view either a single measurement can be reviewed or multiple assays can be selected as a single batch and reanalyzed using a common set of setup parameters. When just a single assay is being reviewed, the screen shown in Figure 4 will appear. This screen can be used to modify the demographics, processing, and calibration associated with a single assay. The Data Review user interface is intended for reviewing the spectra, calibration, and results, and offers an approval process if required by the facility.

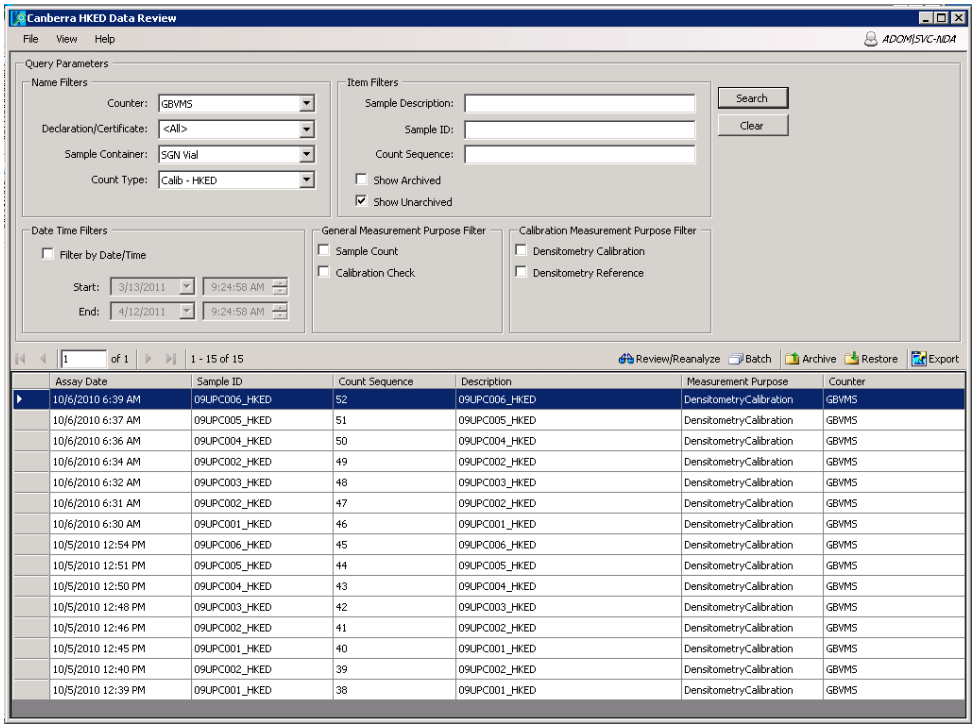

Figure 3: Main view of the Data Review application in Windows-based HKED software

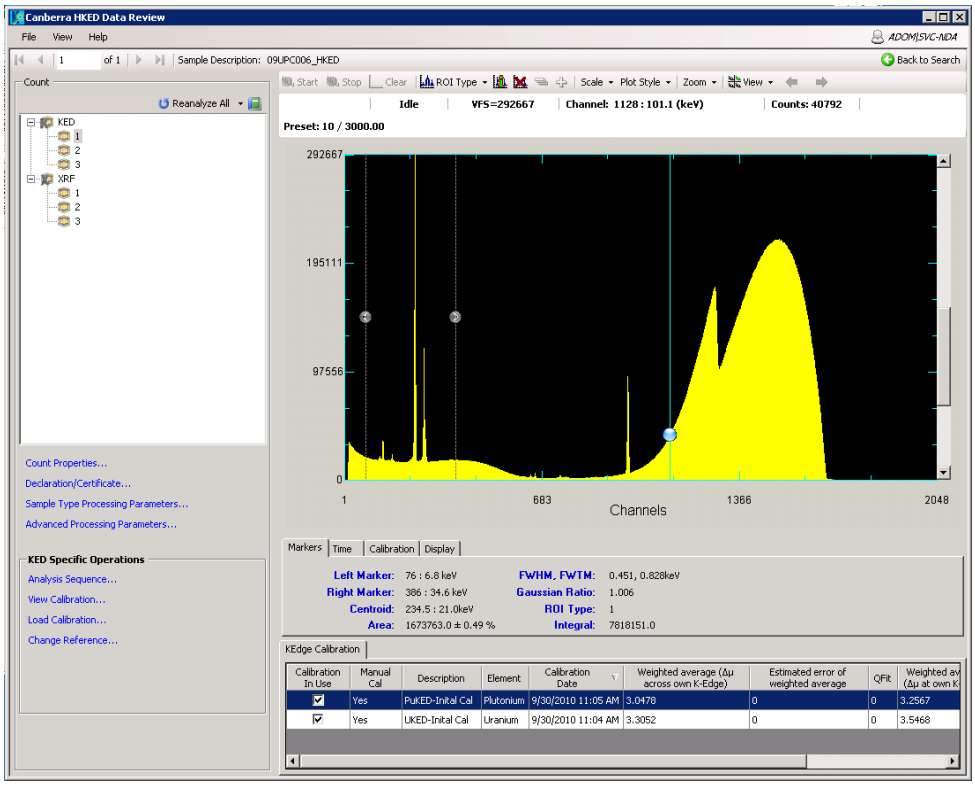

Figure 4: Review/Reanalyze view of the Data Review application in Windows-based HKED software

3. Software validation

The primary goal of the validation of the algorithm porting was to determine if for the identical input values, the identical output values would be achieved. One of the anticipated difficulties of the comparison, given that the two software packages are on two completely different computer platforms, is that rounding differences would preclude identical results. Some rounding is of course inevitable because the two platforms use different internal representations for numbers; in fact the two platforms adhere to different standards. To mitigate this effect the comparison of the analysis was approached in two steps.

The first step was to manually ensure that both software packages were setup with the identical initial parameters. For systems that make the transition from the VMS to Windows software, this is also the easiest approach to transfer the existing calibration parameters on the system from one software to the other. Results of the analysis of the same data using the same initial seed parameter values are shown in the first section below 'Analysis validation'. Here the software runs through the analysis of a sample measurement and produces a concentration result in grams per liter. The second step was to take the same sample measurements and create new calibrations in each of the software packages, thereby creating new calibration parameters. This would be a second approach to transferring the calibration on an existing system from the VMS software to the Windows software, although it is a more involved approach. In this comparison the additional layer of calculations involved could amplify any round-off issues as well as be more subject to any statistical variations between the different samples. In addition, within the original coding of the algorithms not all intermediate calculational parameters are permanently stored in the data. These values are only available as shown in the software user interface or in the printed output reports. In these cases for comparison purposes the value with the greatest precision was chosen, but again inevitably these values had been subjected to some level of round-off. The results of these comparisons is shown in the second section below 'Calibration validation'; this is clearly not a calibration validation in the typical sense, but rather a validation of the calculation of calibration parameters in a comparison of both software packages.

3.1. Validation data

The measured data from two commercial systems were used in the validation study. In this paper these systems are referred as "System 1" and "System 2". Each of the analysis options that are available in the software was tested on both systems. These analysis options include the following:

- Hybrid K-Edge Densitometry (HKED)
- Uranium and Plutonium K-Edge Densitometry (UKED and PuKED)
- Uranium and Plutonium XRF Analysis (UXRF and PuXRF)

The original data was obtained from the commercial systems utilizing the VMS-based software. For each of the systems a number of uranium and plutonium spectra of samples with known concentrations were used to perform calibration and analysis. The calibration process was then repeated with the same spectra, but utilizing the Windows-based HKED software. In order to validate that all analysis algorithms have been properly coded in the new software, the calibration and analysis results were compared to the ones obtained using the VMS-based version.

3.2. Analysis validation

This section presents the Windows-based software analysis validation test results obtained using measured data from two commercial systems. During this step the analysis algorithm were tested to ensure that identical analysis results are produced by both versions of the software when the same calibration parameters are used. In preparation for this step, all calibration parameters available in the VMS system were manually transferred into the Windows-based software. It should be noted that the calibration parameters could only be transferred to a certain precision level, which was limited by the number of significant digits reported in the VMS software for each individual calibration parameter. Therefore, some finite differences in the analysis results could be expected between the two versions of the software.

Note that the complete set of data available for System 1 could be used to test every analysis mode available in the software, whereas for System 2 data were not available on plutonium measurements.

3.2.1. HKED analysis

A total of six reference samples containing dissolver solution with various uranium concentrations were used to perform the Hybrid KED analysis. The uranium concentration spanned the range of approximately 40 to 210 g/L with a U/Pu ratio of approximately 100:1. The HKED analysis results obtained using both versions of the software (Windows-based and VMS-based) are given for System 1 and System 2 in Table 1 and Table 2 respectively. In each table the ratio of the results from the two software packages is shown for the different analysis results. Note that in most cases there was no difference observed in the analysis results between the two versions of the software. In some cases the reported results were not identical, but the difference is clearly at a level that can be attributed to the rounding. The fact that there is no trend in these differences further supports this conclusion.

Sample ID	Uranium Concentration		U/Pu ratio		Plutonium Concentration	
	Value	Uncertainty	Value	Uncertainty	Value	Uncertainty
S1_HKED_001	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000
S1_HKED_002	1.00005	1.00000	1.00000	1.00000	1.00000	1.00000
S1_HKED_003	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000
S1_HKED_004	1.00000	1.00000	0.99992	1.00000	1.00000	1.00000
S1_HKED_005	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000
S1_HKED_006	1.00000	1.00000	0.99992	1.00000	1.00009	1.00000

Table 1: HKED analysis results comparison for “System 1” counts. The table shows ratios between the results reported by a Windows-based HKED software and VMS-based HKED software.

Sample ID	Uranium Concentration		U/Pu ratio		Plutonium Concentration	
	Value	Uncertainty	Value	Uncertainty	Value	Uncertainty
S2_HKED_001	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000
S2_HKED_002	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000
S2_HKED_003	1.00000	1.00000	1.00000	1.00000	0.99994	1.00000
S2_HKED_004	0.99995	1.00000	1.00000	1.00000	1.00000	1.00000
S2_HKED_005	1.00000	1.00000	1.00000	1.00000	1.00006	1.00000
S2_HKED_006	1.00000	1.00000	1.00000	1.00000	0.99992	1.00000

Table 2: HKED analysis results comparison for “System 2” counts. The table shows ratios between the results reported by a Windows-based HKED software and VMS-based HKED software.

3.2.2. UKED analysis

A number of certified samples containing dissolver solution with various uranium concentrations were used to perform the Uranium KED analysis. The uranium concentration range covered by the reference samples for System 1 was from 40 g/L to about 280 g/L and for System 2 from 40 g/L to more than 400 g/L. The UKED analysis results obtained using both versions of the software (Windows-based and VMS-based) are given for System 1 and System 2 in Table 3 and Table 4 respectively. Note that again in most cases there was no difference observed in the analysis results between the two versions of the software. Again for the cases where the reported uranium concentrations were non-identical, the difference is clearly at a level that can be attributed to the rounding.

Sample ID	Uranium Concentration	
	Value	Uncertainty
S1_UKED_001	1.00000	1.00000
S1_UKED_002	1.00000	1.00000
S1_UKED_003	1.00000	1.00000
S1_UKED_004	1.00005	1.00000
S1_UKED_005	1.00000	1.00000
S1_UKED_006	1.00000	1.00000
S1_UKED_007	1.00000	1.00000
S1_UKED_008	1.00000	1.00000

Table 3: UKED analysis results for “System 1” counts. The table shows ratios between the results reported by a Windows-based HKED software and VMS-based HKED software.

Sample ID	Uranium Concentration	
	Value	Uncertainty
S2_UKED_001	1.00000	1.00000
S2_UKED_002	1.00000	1.00000
S2_UKED_003	1.00000	1.00000
S2_UKED_004	1.00000	1.00000
S2_UKED_005	1.00000	1.00000
S2_UKED_006	0.99995	1.00000
S2_UKED_007	1.00000	1.00000
S2_UKED_008	1.00000	1.00000
S2_UKED_009	1.00000	1.00000

Table 4: UKED analysis results for “System 2” counts. The table shows ratios between the results reported by a Windows-based HKED software and VMS-based HKED software.

3.2.3. PuKED analysis

A similar set of measurements was performed on System 1 using reference samples with plutonium solutions. Several samples with plutonium concentration ranging from 36 to approximately 260 g/L were used to validate the Plutonium KED analysis algorithms. As in the case with uranium samples, most of the analysis results were identical for both versions of the software. The comparison results between plutonium concentrations reported by Windows-based and VMS-based versions of the software are presented in Table 5. Only in a single case was there an observed non-identical result, and the difference is clearly at a level that can be attributed to the rounding difference.

Sample ID	Plutonium Concentration	
	Value	Uncertainty
S1_PuKED_001	1.00000	1.00000
S1_PuKED_002	1.00000	1.00000
S1_PuKED_003	0.99994	1.00000
S1_PuKED_004	1.00000	1.00000
S1_PuKED_005	1.00000	1.00000

Table 5: PuKED analysis results for “System 1” counts. The table shows ratios between the results reported by a Windows-based HKED software and VMS-based HKED software.

3.2.4. UXRF analysis

Two sets of reference samples with low uranium concentrations were used to verify the Uranium XRF analysis algorithms. The first set of samples was used on System 1 and covered the uranium concentration range from 5 to approximately 40 g/L. The uranium concentration range of the second set spanned between 0.4 and approximately 9 g/L. This set was measured and analyzed using System 2. In all cases except one the analysis results were identical between the Windows-based and VMS-based versions of the software, as shown in Table 6 and Table 7. The only non-identical result is again visibly at the level of round-off.

Sample ID	Uranium Concentration	
	Value	Uncertainty
S1_UXRF_001	1.00000	1.00000
S1_UXRF_002	1.00000	1.00000
S1_UXRF_003	1.00000	1.00000

Table 6: UXRF analysis results for “System 1” counts. The table shows ratios between the results reported by a Windows-based HKED software and VMS-based HKED software.

Sample ID	Uranium Concentration	
	Value	Uncertainty
S2_UXRF_001	0.99999	1.00000
S2_UXRF_002	1.00000	1.00000
S2_UXRF_003	1.00000	1.00000
S2_UXRF_004	1.00000	1.00000

Table 7: UXRF analysis results for “System 2” counts. The table shows ratios between the results reported by a Windows-based HKED software and VMS-based HKED software.

3.2.5. PuXRF analysis

A total of six reference samples containing plutonium solution, covering a wide concentration range (0.5 to approximately 35 g/L), was used to validate the Plutonium XRF analysis option. Table 8 shows the analysis results, where both versions of the software reported identical values for plutonium concentration.

Sample ID	Plutonium Concentration	
	Value	Uncertainty
S1_PuXRF_001	1.00000	1.00000
S1_PuXRF_002	1.00000	1.00000
S1_PuXRF_003	1.00000	1.00000
S1_PuXRF_004	1.00000	1.00000
S1_PuXRF_005	1.00000	1.00000
S1_PuXRF_006	1.00000	1.00000

Table 8: PuXRF analysis results for “System 1” counts. The table shows ratios between the results reported by a Windows-based HKED software and VMS-based HKED software.

3.3. Calibration validation

This section present the Windows-based software calibration validation results obtained using the same measured data described in section 3.2. The purpose of this test was to verify that if the same calibration measurements and default values for the initial calibration parameters are used, both versions of the software will produce similar calibration results. A wide range of reference measurements were used to create calibrations for different analysis types. As noted previously in this comparison the additional layer of calculations involved could amplify the round-off issues on the two different platforms. Although this approach to transferring a calibration between the two software packages would be more involved, if this approach were taken it would be important to note the effect of such round-off issues.

3.3.1. HKED calibration

The same set of reference counts as described in section 3.2.1 spanning the range from 40 to 210 g/L with a U/Pu ratio being about 100:1 was used. The certificate values were available for each count and were used in the calibration.

Table 9 below shows the major calibration coefficients obtained for the Hybrid K-edge Densitometry analysis mode. Note that there is virtually no difference in FACTBG and FACTPU calibration coefficients obtained using different versions of the HKED software.

Calibration factors	"System 1" counts			"System 2" counts		
	Windows-based HKED	VMS-based HKED	Win/VMS Ratio	Windows-based HKED	VMS-based HKED	Win/VMS Ratio
FACTBG	1.39928	1.39928	1.00000	1.37491	1.37490	1.00001
FACTPU	1.09236	1.09235	1.00001	1.08685	1.08684	1.00001
c1	7.0717E-05	9.4880E-05	0.74533	-9.6182E-03	-9.6659E-03	0.99507
c2	2.1929E-05	1.9597E-05	1.11905	9.8910E-04	9.9390E-04	0.99517

Table 9: HKED calibration comparison results

It can be seen from Table 9 that for the c1 and c2 coefficients there appears to be a significant deviation in the values. These empirical coefficients are used to calculate the non-linearity correction factor for FACTPU in the cases when uranium concentration exceeds 200 g/L. When the different values for c1 and c2 are used to calculate FACTPU, however, the result differs at the level of round-off. This is shown in Table 10 for some concentration values within the calibration range.

Uranium concentration, g/L	FACTPU (Windows-based)	FACTPU (VMS-based)	Win/VMS Ratio
200	1.09236	1.09235	1.00001
205	1.09326	1.09331	0.99995
210	1.09526	1.09526	1.00000

Table 10: Comparison of the FACTPU adjustment factors calculated using c1 and c2 non-linearity coefficients determined by Windows-based and VMS-based software using "System 1" counts

3.3.2. UKED calibration

The same set of reference counts described in section 3.2.2 was used in this comparison as well along with the corresponding certified concentration values for the system calibration.

The calibration factors obtained for the Uranium KED analysis mode are given in Table 11. Note that both calibration coefficients, $\Delta\mu$ (Non_Extrapolated) and $\Delta\mu$ (Extrapolated), determined in the Windows-based software agree well with the same calibration factors determined using the VMS-based software. The difference between the corresponding parameters is clearly at the level that would be expected from round-off.

Calibration factors	"System 1" counts			"System 2" counts		
	Windows-based HKED	VMS-based HKED	Win/VMS Ratio	Windows-based HKED	VMS-based HKED	Win/VMS Ratio
$\Delta\mu$ (Non_Extrapolated), cm^2/g	3.29297	3.29295	1.00001	3.28433	3.28432	1.00000
Estimated error ($\Delta\mu$ (Non_Extrapolated))	0.001187	0.00119	0.99748	0.001339	0.00134	0.99925
$\Delta\mu$ (Extrapolated), cm^2/g	3.52863	3.52862	1.00000	3.51356	3.51355	1.00000
Estimated error ($\Delta\mu$ (Extrapolated))	0.001794	0.00179	1.00223	0.00201	0.00201	1.00000

Table 11: UKED calibration comparison results.

3.3.3. PuKED calibration

The same set of reference plutonium counts as described in section 3.2.3 was used during this calibration.

The calibration factors obtained for the Plutonium KED analysis mode are given in Table 12. Note that both calibration coefficients, $\Delta\mu$ (Non_Extrapolated) and $\Delta\mu$ (Extrapolated), determined in the Windows-based software agree well with the same calibration factors determined using the VMS-based software. Here too the difference between the corresponding parameters is at the level that would be expected from round-off.

Calibration factors	"System 1" counts		
	Windows-based HKED	VMS-based HKED	Win/VMS Ratio
$\Delta\mu$ (Non_Extrapolated), cm^2/g	3.054991	3.05501	0.99999
Estimated error ($\Delta\mu$ (Non_Extrapolated))	0.001503	0.00150	1.00200
$\Delta\mu$ (Extrapolated), cm^2/g	3.259823	3.25984	0.99999
Estimated error ($\Delta\mu$ (Extrapolated))	0.002217	0.00222	0.99865

Table 12: PuKED calibration comparison results.

3.3.4. UXRF calibration

The set of reference counts previously described in section 3.2.4 was used to calibrate the system.

Table 13 contains a summary of the UXRF calibration factors (A0 and A1) obtained on System 1 and System 2 using the different software packages. The difference between the corresponding parameters is at the expected level.

Calibration factors	"System 1" counts			"System 2" counts		
	Windows-based HKED	VMS-based HKED	Win/VMS Ratio	Windows-based HKED	VMS-based HKED	Win/VMS Ratio
A0	3.14878E-02	3.14880E-02	0.99999	3.15488E-02	3.15490E-02	0.99999
Uncertainty A0	3.02657E-05	3.02657E-05	1.00000	6.41737E-05	6.41740E-05	1.00000
A1	1.09754E-04	1.09700E-04	1.00049	-2.59631E-05	-2.59650E-05	0.99993
Uncertainty A1	1.13702E-06	1.13697E-06	1.00005	7.54226E-06	7.54228E-06	1.00000
Covariance (A0, A1)	-2.97901E-11	-2.97900E-11	1.00000	-4.56460E-10	-4.56460E-10	1.00000

Table 13: UXRF calibration comparison results.

3.3.5. PuXRF calibration

The set of reference counts previously described in section 3.2.4 was used to calibrate the system.

Good agreement was also observed in the calibration factors (A0 and A1) with the maximum difference between the corresponding parameters at the expected level.

Calibration factors	"System 1" counts		
	Windows-based HKED	VMS-based HKED	Win/VMS Ratio
A0	4.685978E-02	4.68357E-02	1.00051
Uncertainty A0	3.896271E-05	3.89423E-05	1.00053
A1	1.644716E-04	1.64495E-04	0.99986
Uncertainty A1	1.656417E-06	1.65641E-06	1.00000
Covariance (A0, A1)	-5.833699E-11	-5.83070E-11	1.00051

Table 14: PuXRF calibration comparison results.

3.4. Results of calibrations comparison

In section 3.3 where the calibration parameters were regenerated from the calibration measurement data, there were two cases in which the round-off appeared to affect the resulting parameters more than in other cases. One was in the HKED calibration parameters, where the resulting values for the c1 and c2 parameters appear to differ significantly, but it was shown (Table 10) that the factors that were calculated using these parameters only showed a minimal difference. As a final check, the calibration parameters obtained in each of the Windows- and VMS-based software packages were then used to reanalyze the identical samples, and the resulting concentration obtained in each software package was then compared. This difference was found to be insignificant and well within the performance expected of the instrument. A similar exercise was carried out for the Pu XRF calibration parameters which although were not significantly different, were on average more different than in the other cases. Both these re-analyses were on the samples measured in System 1. The results are shown in the ensuing sections.

3.4.1. Concentration results based on HKED calibration of System 1

Sample ID	Windows/VMS measured concentration ratio			
	Uranium		Plutonium	
	Ratio	Uncertainty	Ratio	Uncertainty
S1_HKED_001	1.0000	0.0028	0.9983	0.0327
S1_HKED_002	1.0000	0.0028	1.0009	0.0289
S1_HKED_003	1.0000	0.0030	1.0003	0.0218
S1_HKED_004	1.0000	0.0028	0.9991	0.0249
S1_HKED_005	0.9999	0.0028	1.0009	0.0220
S1_HKED_006	0.9999	0.0030	1.0002	0.0166

Table 15: Ratio of Windows measured concentration determined using the Windows calibration parameter result to VMS measured concentration determined using the VMS calibration parameter result

3.4.2. Concentration results based on PuXRF calibration of System 1

Sample ID	Windows/VMS measured concentration ratio	
	Ratio	Uncertainty
S1_PuXRF_001	1.0005	0.0028
S1_PuXRF_002	1.0005	0.0027
S1_PuXRF_003	1.0005	0.0027
S1_PuXRF_004	1.0005	0.0031
S1_PuXRF_005	1.0005	0.0037
S1_PuXRF_006	1.0006	0.0061

Table 16: Ratio of Windows measured concentration determined using the Windows calibration parameter result to VMS measured concentration determined using the VMS calibration parameter result

4. Conclusions

Hybrid K-Edge Densitometry (HKED) is a highly accurate non-destructive analysis technique used to determine the uranium and plutonium concentrations in dissolver solutions during fuel reprocessing as an alternative to chemical (destructive) analysis. The technique uses results from two measurements performed simultaneously providing both a K-Edge transmission measurement and an X-Ray Fluorescence measurement of the sample. The HKED Software automates the process of measuring and analyzing samples by controlling the HPGe gamma detectors, and ancillary equipment when needed.

The VMS version of the software was developed more than a decade ago based on the prescription of the original developers for use on computers running VMS operating systems at the time. With increasing concerns for the obsolescence and support of this platform, a new fully industrial, quality-tested PC-platform based software has been developed. The new software has an easy-to-use

graphical user interface for all functions, including setup, calibration, sample measurement, and system maintenance. Although not directly visible to the user, the other benefit of the new software development was in the underlying architecture of the software which allows much more easily the future addition of new analysis algorithms when needed as well as support for new hardware. One of the key goals in planning the transition of the software from the VMS to the Windows platform was in preserving the analysis algorithms, and keeping these unchanged in order to ensure traceability of results.

In this paper the results of the validation tests performed with the new Windows-based HKED software showed that the core analysis algorithms remain unchanged and that they and the supporting software will provide the same level of accuracy as implemented in the original VMS-based version of the software.

5. References

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Quench gas and shaper/discriminator selection influence on ^{3}He tube performance for spent fuel applications

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LA-UR 11-02641

Abstract:

Current ^{3}He tubes utilized in neutron coincidence counting use different quench gas admixtures to shorten the avalanche process. In addition amplifier modules with different shaping characteristics are used to process detector signals. Both of these aspects affect the detector response. In the current paper, ^{3}He tubes with several quench gas admixtures (CO_2, N_2, Ar+CH_4 and CF_4) and amplifier modules (PDT, AMPTEK, BOT) are compared. The plateau characteristics, gamma-sensitivity and dead-time of individual counters in combination with the listed amplifier modules are compared to determine optimum amplifier module/counter performance for the spent fuel applications.

Keywords: plutonium content, spent fuel, ^{3}He count rate capabilities, NDA

1. Introduction

The Next Generation Safeguards Initiative (NGSI) of the U.S. DOE has initiated a multi-laboratory/university collaboration to quantify the plutonium (Pu) mass in, and detect the diversion of pins from, spent nuclear fuel assemblies with non-destructive assay (NDA) techniques. The goal of this research effort is to improve the technology base of safeguards.

Safeguarding and determination of plutonium mass in spent fuel assemblies represents a very challenging task due to high gamma and neutron contributions from accumulated fission products and actinides that mask the fissile content signatures. The surface gamma-ray dose of a typical spent fuel assembly can reach levels as high as 10^5 R/hr. Use of lead shielding can reduce the dose to 1-10 R/hr at the detector face, which still represents levels not typically encountered in traditional safeguards. Thus a detection system with low gamma sensitivity is desirable. In addition a fast detection system is required, capable to handle rates of the order of 10^6-10^7 counts per second. Recently developed Differential Die-Away Self-Interrogation (DDSI) technique [1] aims to provide plutonium mass information based directly on the measurement of spontaneous fission neutrons from ^{244}Cm and induced fissions in the sample. The technique utilizes the doubles/singles ratio and the early and late counting gates to determine the fissile content from the ratio of count rates in these two gates. This ratio is subsequently related to plutonium mass using weighted contributions from three major fissile isotopes (^{235}U, ^{239}Pu and ^{241}Pu). The need to provide a good separation between the early and late gates in the DDSI technique together with high count rate requirements places tight limits on timing characteristics of neutron detection system.

Traditional neutron coincidence counting utilizes ^{3}He based proportional counters, which are favoured for their high stability, efficiency of neutron detection and low gamma sensitivity. Nevertheless, in order to operate a ^{3}He based counter in the demanding conditions such as spent fuel applications, its gamma sensitivity and count rate capabilities need to be optimized. In the current investigation, we focus on the influence of different types of quench gas and signal processing electronics on the performance of the ^{3}He based system.

The primary role of quench gas is to suppress photo-emission caused by de-excitation of atoms of the fill gas and related secondary electron emission. In addition presence of quench gas molecules contributes to increase in electron drift velocities and increase of the stopping power of $^{3}He(n,p)^{3}H$ reaction products. Different types of quenching molecules are used based on application needs. The most typical in neutron coincidence counting applications is an $Ar+CH_4$ quench gas due to its favourable count rate characteristics [2]. However, due to the high *Z* of Ar, it is more sensitive to gamma interactions [3]. In addition CO_2 or N_2 quench gas admixtures are frequently used, which were shown to be capable to operate in increased gamma backgrounds with similar performance [4]. Nevertheless, N_2 quench gas admixture results in longer pulses, which can limit its count rate capabilities. A use of CF_4 quench gas was suggested as a possible candidate to exhibit lower gamma-ray sensitivity [5] and higher electron drift velocity [6], which are favourable characteristics for spent fuel applications.

The signal processing electronics can be optimized to achieve desired signal characteristics and count rate capabilities for given application needs. The type of shaping and shaping time constants will influence the gamma pileup rejection as well as count rate capabilities of the system. Short shaping time constants result in faster decay times of the detected pulses and allow the system to be operated at higher count rates. Additionally short shaping times reduce contribution of gamma pileup relative to neutron pulses. The most frequently used signal processing electronics in combination with ^{3}He-based counters are AMPTEK and Precision Data Technology (PDT) amplifier modules. In addition the BOT Inc. amplifier modules are considered for safeguards applications by IAEA.

In order to evaluate capabilities of different ^{3}He proportional counters for spent fuel applications we performed a dedicated experimental evaluation of ^{3}He counters with several different quench gas admixtures (CO_2, N_2, $Ar+CH_4$ and CF_4). Each ^{3}He counter was combined with typical amplifier modules (PDT, AMPTEK, BOT). The high voltage (HV) plateau, gamma sensitivity and dead-time characteristics of individual counters in combination with the listed amplifier modules were evaluated.

2. Experimental evaluation

Four ^{3}He counters with CO_2, N_2, $Ar+CH_4$ and CF_4 quench gas admixtures were evaluated. Each counter was 12” long with 1” diameter and fill gas pressure of 4 atm. Each counter was combined with standard AMPTEK-A111, PDT-10A and BOT amplifier modules. The AMPTEK-A111 amplifier module has bipolar shaping with very short time constant of the order of 190 ns. PDT amplifier module is available in many different application specific versions. In current testing the PDT-10A version with bipolar shaping and shaping time of ~500 ns was utilized. This PDT is used in many ^{3}He based neutron coincidence counters and thus serves as a good performance reference. The BOT amplifier module was designed for use with wide variety of proportional counters. The unit used in the reported test activity provides a unipolar shaping with 2.5 μs shaping time. All of these units provide user-adjustable gain and threshold. In the current test activity the gain of each amplifier module was adjusted to correspond to an optimum performance of a CF_4 admixture in the ^{3}He tube.

During the measurement each counter was placed in a cylindrical polyethylene block and irradiated by a 98 μCi ^{252}Cf source. Each counter/electronics combination was tested for the shape of high voltage plateau, gamma sensitivity and dead-time. During the HV plateau and gamma sensitivity measurements the digital output of amplifier module was recorded using JSR-12. To evaluate system dead-time the digital output of amplifier module was recorded in list mode to allow for data recording on event-by-event basis. Using this information a time interval analysis was utilized to extract dead-time information for each counter/electronics combination. To record data in list mode, a List Mode Multiplicity Module developed at LANL was used [7].

2.1. High voltage plateau

High voltage plateau was measured for each counter/electronics combination in the range of 1200 V – 2000 V in 20 V increments. Based on this information, an operating voltage for every system was determined as 40 V into the plateau, above the knee. The HV plateau results for the four quench gas admixtures and tested amplifier modules are summarized in Figure 1.

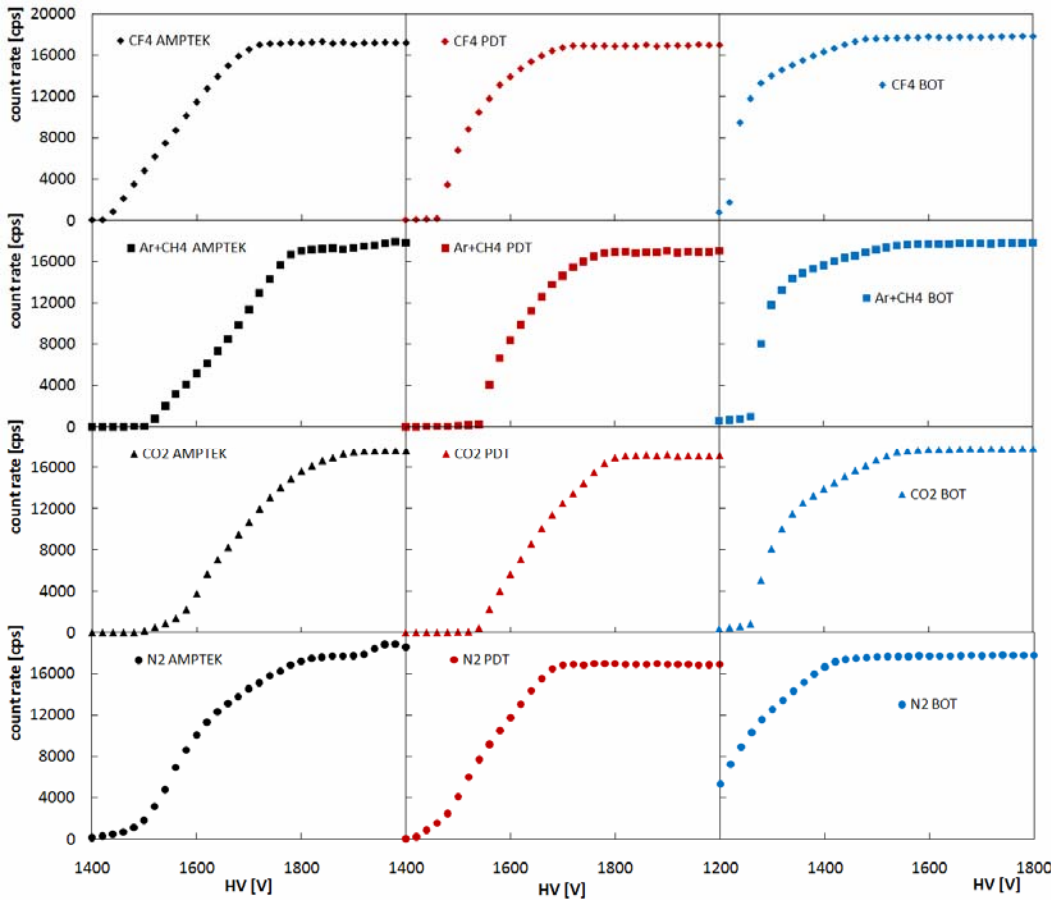

Figure 1: Comparison HV plateaus measured using ^{3}He counters with Ar+CH_4, CF_4, CO_2 and N_2 quench gas admixtures. Results were acquired using AMPTEK-A111 (left), PDT-10A (middle) and BOT (right) amplifier module. Note different x-axis in case of BOT amplifier module.

Differences between the quench gas admixtures are clearly visible in Figure 1. A very long and stable plateau is observed in case of CF_4 quench gas for all the amplifier modules used in the test. The slope of the plateau is better than 0.5 %/100 V. The Ar+CH_4 as well as N_2 quench gas admixtures, when connected to AMPTEK-A111 amplifier module, exhibit an onset of a second hump on the plateau. This hump can be attributed to multiple triggering of electronics on a single neutron capture event. Variation of ionization tracks traversing ^{3}He counter translates into complex structures present in the detector current pulses. Due to the short shaping time in the amplifier the discriminator can retrigger on these structures formed in a single event. This effect is more pronounced in case of N_2 gas. The operational high voltage is normally set below the level where this effect becomes important. As can be seen from comparison with PDT and BOT amplifier modules the second hump is removed when longer shaping time electronics is used.

Figure 1 also illustrates influence of different signal processing electronics on the shape of HV curve. A well defined plateau is observed for all the quench gas admixtures combined with PDT-10A or BOT electronics. As noted above, fast shaping time of the AMPTEK-A111 affects the response of Ar+CH_4 and N_2 quench gasses by additional hump on the plateau. In addition the shape of the HV plateau is no longer well defined in case of N_2 quench gas and only very short plateau is observed in case of CO_2 admixture when combined with AMPTEK amplifier module. This is a consequence of interplay between the short time constant of the AMPTEK shaper and longer collection time of these quench gas admixtures. Thus use of AMPTEK amplifier module with these two quench gas admixtures is not advisable.

Use of different amplifier modules also affects the extent of the HV plateau and shape of the slope of the initial count rate increase with increasing HV. These differences can be attributed to the differences in the types and time constants of the shaping electronics. The shaping type and time constant of an amplifier influences the signal amplitude and thus the shape of a pulse height spectrum. With shorter shaping times, a smaller portion of the input charge pulse is integrated

resulting in incomplete energy information. The short shaping time causes deterioration of full energy peak in the pulse height spectrum and increased contribution of low energy pulses. With increasing shaping time, more complete charge integration is performed, which allows preserving complete energy information for sufficiently long shaping. The differences in the shapes of the pulse height spectra translate into the HV plateau as observed in Figure 1. The long shaping time of BOT amplifier module allows for preserving the most complete energy information, thus a sharp initial increase in count rate with HV followed by a smooth transition into the plateau region is observed.

2.2. Gamma sensitivity

To evaluate the gamma sensitivity of each counter/electronics combination measurements were performed using ^{137}Cs sources of ~450 mR/hr gamma dose rate at the detector face. The combined dose rate of the ^{137}Cs sources is the maximum dose rate available under our laboratory conditions. The HV curve results are compared to bare neutron source data in Figure 2.

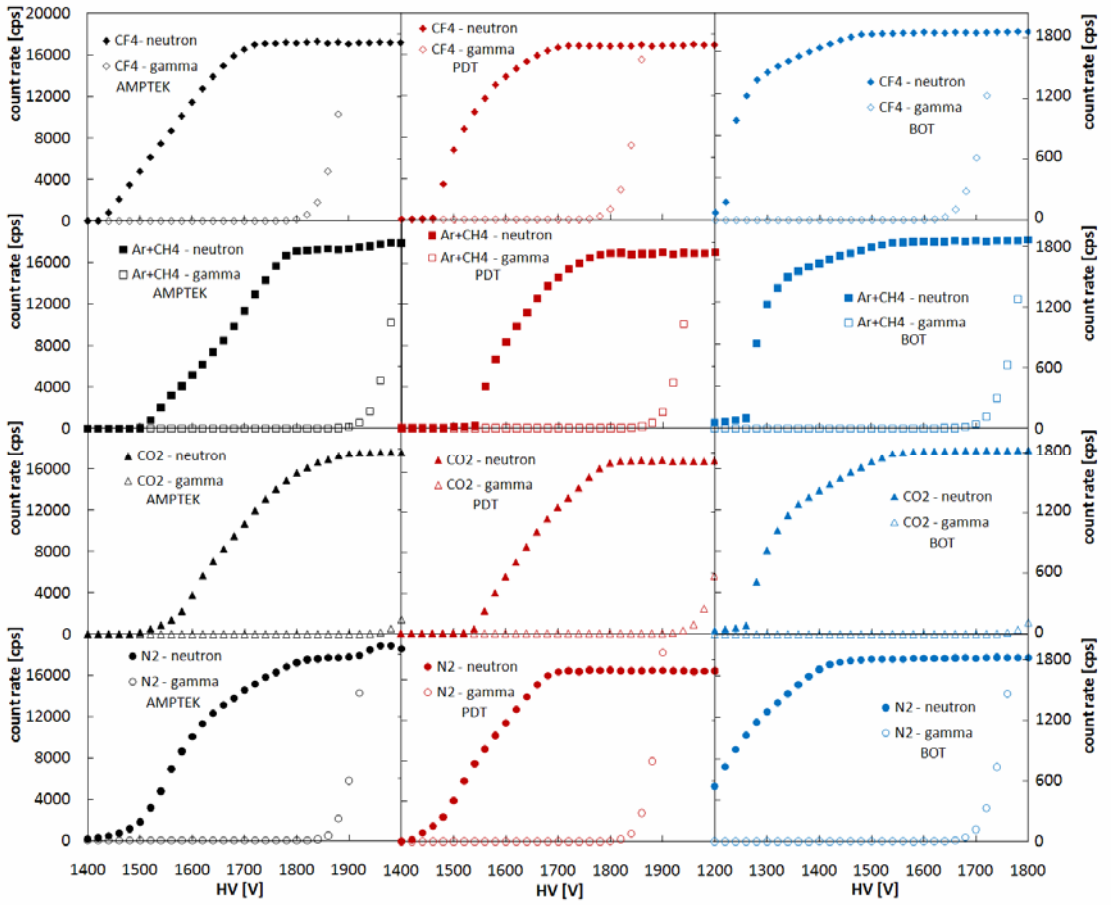

Figure 2: HV plateau measured with ^{252}Cf source (full symbols) and using ^{137}Cs sources of ~450 mR/Hr dose rate at the detector face (open symbols) for all the quench gas admixtures and electronics combinations investigated. Note different scale on x-axis in case of BOT amplifier module. The count rate corresponding to gamma events is shown over an expanded range (right vertical axis).

An increase of count rate due to gamma-ray pileup is clearly visible in Figure 2 for all the quench gas admixtures. Comparing different quench gas admixtures it can be seen that CO_2 and N_2 combined with BOT electronics result in the longest plateau before an onset of gamma pileup, which corresponds to ~ 200 V. The length of the plateau in case of Ar+CH_4 and CF_4 admixtures is slightly reduced and corresponds to ~ 160 V. The onset of gamma pileup is similar for all the quench gas admixtures when using PDT amplifier module. The length of the plateau in this case corresponds to ~ 120 V in case of CO_2 and N_2 and to ~ 100 V in case of Ar+CH_4 and CF_4 quench gas admixtures. A very short apparent plateau before an onset of gamma pileup is observed for CO_2 and N_2 quench gas admixtures combined with AMPTEK amplifier module. This is a consequence of an earlier observation that the use of the AMPTEK amplifier module does not provide good plateau characteristics for these

gas admixtures. The gamma sensitivity of Ar+CH_4 and CF_4 quench gas admixtures when combined with AMPTEK amplifier module is similar as in case of PDT electronics.

Due to the higher *Z,* Ar gas is expected to be more sensitive to gamma pileup than lighter CF_4 molecule. The similar gamma sensitivity observed for of both quench gas admixtures suggests that the effect is dominated by the interactions in the counter walls and fairly insensitive to the type of quench gas.

2.3. Dead-time

Another parameter crucial to achieve high count rate capabilities is the system dead-time. Dead-time is a complex function of the system parameters and is affected by the properties of the counter (such as detector fill gas or applied HV) as well as by signal processing electronics (such as shaping time and type of shaping). Use of list mode data acquisition allows a direct investigation of dead-time in the detection system. Dead-time information can be extracted from time interval analysis of list mode data, where frequency of occurrence of a given time interval between two subsequent pulses is evaluated.

Dead-time was evaluated for each counter/electronics combination using a ^{252}Cf source. Comparison of time interval distributions for the four investigated quench gas admixtures in combination with AMPTEK-A111 and PDT-10A amplifier module is shown in Figure 3 (left) and (right), respectively. Comparison of influence of signal processing electronics on time interval distribution is shown in Figure 4.

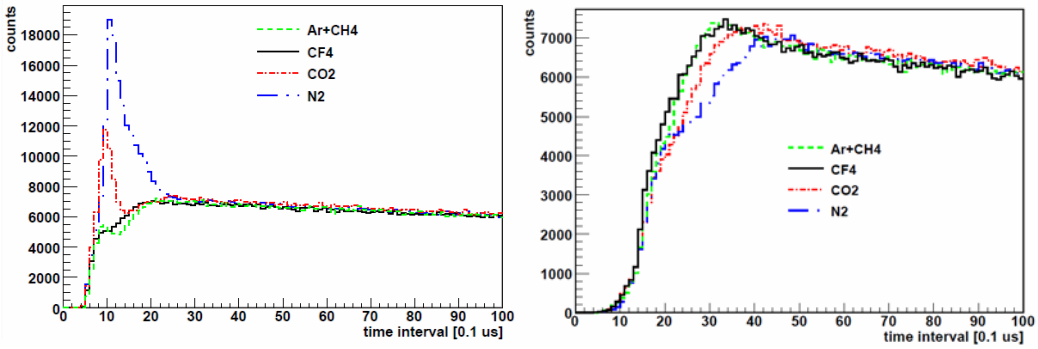

Figure 3: Time interval analysis results for Ar+CH_4, CF_4, CO_2 and N_2 quench gas admixtures with AMPTEK-A111 (left) and PDT-10A (right) amplifier modules.

Figure 3 provides an overview of influence of different quench gas admixtures on the system dead-time. It can be seen that the minimum interval before a second pulse can be detected, which represents system dead-time, corresponds to ~ 600 ns and ~ 1.6 µs in case of AMPTEK and PDT electronics, respectively. This interval is largely independent of the type of quench gas. Nevertheless, the short time-interval range in case of AMPTEK amplifier module is dominated by a pronounced spike in case of CO_2 and N_2 admixtures. However, much less pronounced, structure is apparent in the case of Ar+CH_4 quench gas. This spike is a consequence of the interplay of short shaping time of the AMPTEK amplifier module and different charge collection times from the investigated quench gas admixtures.

The products of neutron interaction in the ^{3}He gas can traverse the proportional counter in various directions causing spread in the rise time of charge pulses from the detector. The shortest rise times correspond to ionization tracks parallel to anode wire. The charge pulse spread is transformed into variation of shapes and amplitudes of current pulses, which can reveal complex structures with multiple humps. These structures are apparent in particular in case of tracks oriented perpendicular to the anode wire. Due to the short time constant of AMPTEK shaping amplifier, its output pulse is dominated by the structures present in the detector pulse. Therefore a re-triggering of discriminator on the complex pulse structures can be expected, which causes false events. The spike on the timescale of approximately 1 µs observed in Figure 3 (left) is a consequence of this effect. The importance of discriminator re-triggering can be eliminated by use of a longer shaping time as shown in Figure 3 (right) in case of PDT-10A amplifier module with shaping time of ~500 ns. In addition the effect is significantly reduced in case of CF_4 admixture due to fast electron drift velocities in this quench gas.

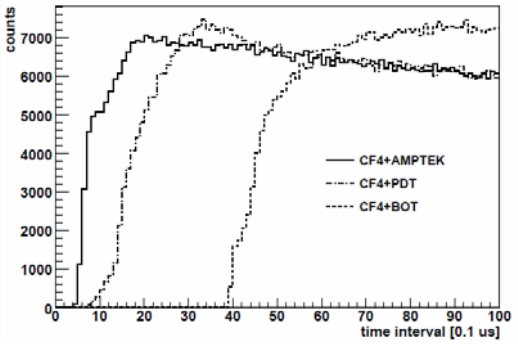

Figure 4: Time interval analysis for ^{3}He counter with CF_4 quench gas and different signal processing electronics.

Figure 4 shows a comparison of time interval distributions for CF_4 quench gas and different signal processing electronics. A clear effect of signal processing electronics on the system dead-time can be seen from the figure. The shaping characteristics and short shaping time of AMPTEK result in the fastest system response with shortest dead-time of the order of 600 ns. The longer shaping time characteristics of PDT-10A amplifier module cause a slower system response with dead-time closer to ~ 1.6 µs. The tested BOT amplifier module with internal shaping time of 2.5 µs results in the longest dead-time of the order of 4 µs. Figure 4 illustrates importance of optimizing the electronics for the high count rate capabilities. From the tested amplifier modules, AMPTEK in combination with CF_4 quench gas provides the most favourable timing characteristics. Nevertheless, the shaping time constants of PDT and BOT amplifier modules can be customized by manufacturer to allow shorter shaping times.

3. Discussion and conclusions

Results of the quench gas/electronics comparison presented in this paper clearly indicate the importance of optimization of the type of quench gas and signal processing electronics for spent fuel applications. In the spent fuel applications, detectors will be exposed to extreme environments not encountered in typical safeguards use for which existing techniques were optimized. Specifically improvements in gamma-ray sensitivity and count rate capabilities are highly desirable. As noted earlier, the use of CF_4 quench gas was suggested as a possible alternative to provide lower gamma sensitivity and faster response than other traditionally used quench gas admixtures [4,5]. In the current investigation a 4 atm ^{3}He counter with CF_4 quench gas admixture was evaluated against other typical quench gas admixtures. The concentration of CF_4 quench gas was optimized to provide comparable gain to a typical counter utilizing Ar+CH_4 quench gas.

The comparison of HV plateau from investigated quench gas admixtures showed that the CF_4 quench gas exhibits a very stable (< 0.5 %/100 V) and long plateau. Contrary to N_2 and Ar+CH_4 admixtures no double counting increase was observed in case of CF_4 quench gas. Considering the high heat output of a typical spent fuel assembly, the stable plateau of CF_4 suggests a very good long term and environmental stability can be expected in case of this counter. Gamma sensitivity of the investigated counters was evaluated at app. 450 mR/hr dose rate at the detector face – the maximum dose rate available under our laboratory conditions. It was found that the ^{3}He-CF_4 gas exhibits very similar gamma sensitivity as ^{3}He-Ar+CH_4 gas despite the high *Z* of argon. This observation is likely a consequence of gamma interactions in the counter walls. Thus, it can be expected that in high gamma fields ^{3}He-CF_4 gas fill will exhibit similar behaviour as ^{3}He-Ar+CH_4.

As shown in Figure 5, the signal processing electronics has a strong influence on the system dead-time and thus on the achievable count rate capabilities. By selecting a faster electronics time constant, the energy resolution of the system may be compromised. However, in neutron coincidence counting only arrival times of individual pulses are recorded and used to determine sample parameters based on observed correlations. In the case of spent fuel applications, the high count rate capabilities are paramount for successful measurements. Thus, the selection of the signal processing electronics for spent fuel applications need to reflect these requirements. As shown in the current investigation, the choice of electronics has a strong influence on the system timing characteristics. Short time constants of the shaping electronics clearly result in shorter dead-times. Combination of fast timing in electronics

with fill gas with fast electron drift velocity/short ion track length results in the most favourable timing characteristics as seen in case of CF_4 quench gas.

Overall, the use of ^{3}He-CF_4 filled counter results in very long and stable plateau allowing for stable operation under varying environmental conditions. In addition the use of CF_4 quench gas and AMPTEK amplifier module with short time constant provides the best timing characteristics of all the systems investigated. Thus, although the gamma sensitivity of CF_4 quench gas is comparable to Ar+CH_4, its short dead-time and very good stability make it the most suitable choice for the spent fuel applications among the investigated quench gas/electronics combinations.

5. Acknowledgements

The authors would like to acknowledge the support provided by the Next Generation Safeguards Initiative (NGSI), Office of Nonproliferation and International Security (NIS), National Nuclear Security Administration (NNSA) for this research. Authors would like to express their thanks for fruitful discussions with Dr. Kiril Ianakiev.

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Proposed Simulation of KED Densitometry and K-XRF Spectra for the Hybrid K-Edge Densitometer Instrument

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Abstract:

The Hybrid K-Edge densitometer instrument is utilized for safeguards measurements to determine the SNM concentrations of dissolver and product solutions. The dissolver solutions can be quite complex due to varying concentrations of SNM, associated minor actinides, and the presence of fission products. The complexity increases as medium- and high burn-up (BU) materials are engaged. Efforts to design and implement algorithms to resolve these complex situations are somewhat impeded by the lack of available challenging KED and K-XRF data. We are developing a simulator program, SIMHKED, which can generate KED and K-XRF detector spectral information based on a large dynamic range of SNM concentrations and for any level of BU for specific fuel assemblies. Additional design goals of SIMHKED are to input the measured detector responses and reference spectra and the option to compute the detector responses and reference spectra using standard Monte Carlo techniques. A discussion of the techniques and algorithms we intend to utilize within SIMHKED as well as the preliminary program flow are provided.

Keywords: K-Edge; densitometer; KED; XRF; simulate; EDXRF

1. Introduction

The Hybrid K-Edge (HKED) instrument [1] is generally utilized (Rokkasho, Sellafield, La Hague) in safeguards and process control applications where a range of heavy metal (U, Pu) dissolver and product solution concentrations, associated with the reprocessing of spent nuclear fuel, are quantified. The HKED technique is a Non-Destructive Assay (NDA) technique used as an alternative to Isotope Dilution Mass Spectrometry (IDMS) which is a Destructive Assay (DA) technique.

The HKED instrument consists of a K-Edge (KED) densitometer in conjunction to Energy Dispersive X-Ray Fluorescence spectrometer (EDXRF). The KED provides sensitivity to U and Pu in the concentration range 30 g/l to greater than 400 or 500 g/l. The EDXRF operates in the K-Edge region producing K-XRF spectra and is sensitive to heavy metal (HM) concentrations in the range of 500 mg/l to 30 or 40 g/l. Accuracy sensitivities to as low as 0.25% can be expected for the HKED instrument

There are two basic types of HKED instruments depending on the sample vial type. The single sample vial (SSV) and the composite sample vial (CSV). The SSV has a single position for the KED and K-XRF measurements whilst the CSV has two separate sample positions; one for the KED and the other for the K-XRF. The SSV is more suitable for incorporating a sample changer whilst the CSV has a better geometry for facilitating a third detector, *i.e.*, a high-resolution gamma spectrometer (HRGS). Although the geometries and detector responses are somewhat different between the SSV and CSV the hybrid analysis is the same.

1.1. HKED Analysis

The crux of the hybrid analysis [1] is to evaluate the major heavy metal (HM) element with KED and then obtain the ratio of the minor element from the K-XRF. The K-XRF analysis is Region-Of-Interest (ROI) based and has limitations when interfering elements, *e.g.*, UKα1 and PuKα2 are engaged. In practice this is not an issue since the algorithms were designed for 100:1 U:Pu (PWR fuel assemblies) and the peak ROI is generally a single FWHM. Not such the case when MOX 1:1 product solutions are encountered.

If the major element is less than a certain concentration, *e.g.*, 30 g/l, then a K-XRF ROI analysis is performed where the absolute yield of the associated Kα or Kβ is calibrated as a function of matrix and HM concentration.

1.2. Necessity for Simulated HKED Data

The biggest issue that exists today for CHKED analysis is the general requirement that a passive spectrum be performed prior to any input solution so as to account for excitation due to photons emitted from fission products (FP). This measurement of a passive spectrum is time consuming and costly, if not logistically impossible, since the passive assay count time may be 5 to 10 times longer than the active. Attempts [1] have been made to model the excitation due to FP's using FP signatures in the active measured data, however, the results have not proven to be worthy or acceptable. The FP problem is compounded since the FP's are generally short lived and spent fuel cooling times and process control delay times alter the fission product photon flux as a function of time which essentially negates any attempt to produce some sort of FP correction term from the active assay alone.

If simulated HKED data is generated for any level of burn-up (BU), spent fuel cooling time, and process control delay time then perhaps new FP correction algorithms may be developed. This may also require the simulation of High-Resolution Gamma Spectroscopy (HRGS) to provide the MeV-range fission product spectrum, in conjunction to the KED and K-XRF, for investigation purposes. For example, it is known [2] that for PWR assemblies the ratio Cs-134/Cs-137 ratio is an indication of the BU and that the FP photon yield as a function of cooling time is also known [3].

As the nuclear fuel cycles progresses and advances, and the level of BU increases, the inclusion of the minor actinides (MA) Am, Np, and Cm are to be considered and perhaps may have significant quantities (SQ) assignments [4]. Although the accepted hybrid analysis technique [1] has provisions for quantifying Am, it only does so when both U and Pu are present. There are no standard analysis provisions for stand-alone Am as well as for Np and Cm.

The current analysis standard [1] was designed and tested for the PUREX reprocessing technique. It is not completely clear as to all the challenges that the pyrochemical reprocessing method might present such as the inclusion of significant matrix effects. Generated simulated HKED data based off the pyrochemical reprocessing technique would be certainly beneficial for validating HKED instrument for such processes.

The radiolysis effects [5] on the sample jugs observed from relatively high Pu concentrations (due to the α-decay) can produce biases if the appropriate evaporation correction is not applied. Including in the simulation radiolysis effects by simulating the delay after the sample was jugged.

For any given assay it may be discovered that the X-ray HV or beam current may have been improperly set or that the W target for the X-ray tube has suffered a substantial amount of degradation. All of these processes can lead to a shift in the KED endpoint energy and/or some shift in the shape of the X-ray continuum. This can lead to biases in the U/Pu ratio since the Pu X-rays are significantly closer to the endpoint energy than the U X-rays [5]. Simulations can reveal the correction that is required once such situations are engaged.

The creation of simulated K-XRF data will allow the Investigation of the necessity of fitting the Ge escape peaks [6] and how not doing so may introduce biases when other actinides are introduced.

Training for the HKED instrument is often performed in the classroom since the HKED instrument is in a radiation area and may not be logistically accessible. Having the ability to generate simulated data in the classroom would certainly be beneficial and provide “real” experience.

On occasion, within the On-Site Laboratory (OSL), the purified waste is examined in the HKED instrument to see if there is any presence of U or Pu. The standard algorithms [1] cannot handle such complex spectra and the resultant data is merely inspected for the presence of any U or Pu X-rays. Simulating purified waste spectra will be extremely beneficial in determining the necessary algorithms and detection limits that would be engaged.

It is clear that the development of simulated HKED data with provisions of an easily adjustable geometry composer will aid in the design for future, more optimized, HKED instruments.

Other constantly engaged issues such as small errors in sample positioning and detector gain shifting can be simulated so as to help explain certain biases when encountered. For example, an examination of a QC chart may display inexplicable, day to day, biases which may be well explained with simulations.

2. XRF Simulations

The most difficult and daunting task is simulating the K-XRF spectra since the models must consider many effects, *e.g.*, coherent and incoherent scattering and K series excitation functions. We have found three independent references [7,8,9] related to generating K-XRF spectral data applicable to the HKED instrument.

2.1. SIMXRF

The SIMXRF application [7] generates an incoherent K-XRF spectrum supporting Th, U, Pu, Am, Np and Cm directly from measured reference spectrum adjusted for X-ray tube current and measurement time. The X-ray peaks, Kα1-3, and the five most prominent Kβ’s are simply superimposed, based on computed peak areas, using standard X-ray response function algorithms with appropriate background.

The superimposed X-ray peak areas are therefore dependent on the relative declared concentrations, average element atomic mass, relative detector full-energy peak efficiency, simulated live time, X-ray generator current, the integrated attenuation-corrected peak excitation integral, and the peak energy calibration factor. The peak energy calibration factor is utilized to normalize the simulation to real calibration data. As a good approximation, the incoherent scatter cross-section for the peak energy can be substituted for the excitation integral.

2.2. NASU

The approach by Berlizov [8] accomplished while at the National Academy of Sciences of Ukraine (NASU) is the most comprehensive means for generation K-XRF spectra, however, it does not produce the densitometer (KED) spectra. The K-XRF is accomplished in three basic steps: the first step models the X-ray source target; the second step models the interaction of the photon flux within the solution; and the final step is the detection of the fluoresced X-rays engaged by the XRF detector. Using MCNP, the three processes are modeled in a probabilistic manner incorporating the instrument geometry. The first two steps are separated from the last step (detection of fluoresced X-rays) where the detector response to the fluoresced X-rays may be computed or derived from measurements (or ISOCS).

The model for the X-ray tube was based on a Tungsten target situated at 23 degrees with a 1 mm Be window. The instrument geometries supported were for both the single sample vial (SSV) and the composite sample vial (CSV) as well as primary beam filter of Cd of 1 mm thickness and stainless steel of 0.5 mm.

The X-ray spectral flux was shown to have vertical asymmetries (+/- 4%) with higher energies at the bottom and lower energies near the top most significantly increasingly worse at 150 keV. The horizontal direction, as expected was, generally less than 1% and appears to be independent of energy.

Bremsstrahlung model greater than the W x-rays was shown to be in good agreement with experiment. The model was used for tabulating the bremsstrahlung flux within the MCNP models utilized for the HKED instrument. Additional source biasing was elected when generating the bremsstrahlung photon flux for energies at the U K-edge up to the endpoint energy.

With source biasing it was observed that 10^7 – 10^10 excitation photons are required to generate a single fluoresced X-ray. To counteract this, several variance reduction MCNP options, *i.e.*, Point Detector, Geometry Splitting/Russian Roulette, Forced Collisions and Selective Scoring were utilized bringing the computation time from 10^6 to 10^7 minutes to just under 10 minutes with 0.5% RSD at 3 g/l for UKα1 and PuKα1. No noticeable biases were introduced utilizing the MCNP reduced variance options.

Coherent scattering was omitted from the photon transport model, due to the selection of the Point Detector reduced variance MCNP option and caution was expressed in that this might introduce biases as much as 0.5% in the UKα1/PuKα1 peak ratios. It was also discovered that differing MCNP libraries for photo-atomic data, *i.e.*, MCPLIB, MCPLIB02, MCPLIB03 and MCPLIB04 could differ as a shift by as much as 0.5% in the UKα and PuKα, however, the relevant ratios were consistent. This was most notable for MCPLIB04 when compared to the older libraries.

Doppler broadening was also emitted since the tests showed no reasonable effect.

One serious drawback of using MCNP is the way that Kβ lines, only two considered, are weighted averages of Kβ1-Kβ3-Kβ5 and Kβ2-Kβ4. The author does acknowledge this issue and recommends the photo-atomic library (MCPLIB) be appropriately modified.

The NASU model was utilized to generate several U:Pu K-XRF spectra in the range 3% to 17.4% for real samples and the results showed very good agreement (less than 0.5%) with the measured UKα1/PuKα1 ratio when the proper X-ray peak response function fitting SW was applied.

The model also was applied to situations were other matrix elements, *e.g.*, those introduced in the pyrochemical processing of spent fuels where the interfering matrix element can be much higher in

concentration the U and Pu. Experimental validation of the applied matrix correction factor for Bi over the concentration range 0 – 200 g/L was supplied.

2.3. NCSU

Researchers at North Carolina State University (NCSU) have developed [9] a photon transport Monte Carlo approach for EDXRF applicable to generating K-XRF spectra similar to other, available, Monte Carlo codes as shown in Figure 1. The approach, called **CEARXF5**, appears to include all the X-Ray modeling requirements of **MCNP5**, such as variance reduction but includes all atomic shells, specifically, all the Kβ's, unlike **MCNP5**.

The author states that the input geometry configuration and definition is exactly the same as that for MCNP5. The **CEARXF5** package uses the Lawrence Livermore National Laboratory (LLNL) Evaluated Photon Data Library (EPDL) and Evaluated Atomic Data Library (EADL) resources. The EPDL provides the photon transport and the EADL provides the atomic relaxation (fluorescence) data.

Although the ultimate goal of the author is to produce a simulated XRF spectrum to assist in deconvolving the complex X-ray spectra from a particular measurement, it appears that the CEARXRF5 code might be applicable in generating K-XRF spectra for the hybrid K-edge.

CODE	CEARXRF 5	EGS4	ITS 3.0	MCNP 5
Establishment	NCSU, USA	SLAC, USA KEK, Japan NRCC, Canada	SAND, USA	LANL, USA
Particles	Photon	Photon/Electron	Photon/electron	Neutron/Photon/electron
Elements(Z)	1-100	1-100	1-100	1-94
Energy Regime	1kev-1 MeV	1kev – 100Gev	1kev-100Gev	1kev-100Gev
XRF Physics	All shells.	$K_{\alpha 1}, K_{\alpha 2}, K_{\beta 1}, K_{\beta 2}$ and L	All K and L, Average M and N	$K_{\alpha 1}, K_{\alpha 2}, K_{\beta 1}, K_{\beta 2}$ and average L
Photon Physics	PE, Incob, Coh, Doppler, Polarization	Same + Pair	Same-Doppler Polarization	Same – Polarization
Geometry	General	General	General	General
Variance Reduction	Powerful	Basically analog	Few and simple	Powerful for transport analog for spectra
Correlated Sampling	Yes	No	No	Yes (from 4B)
Library spectra	Yes	No	No	No
Differential Operators	Yes	No	No	No
X-ray Coincidence simulation	Yes	No	No	No

Figure 1 Comparison[§] of features between several photon transport Monte Carlo codes.

[§] Reproduced from reference [9].

3. KED Simulations

Simulation of the KED is generally straight forward since the inverse of the Generalized KED [10] can be simply applied, as a first approximation, to an analytically generated bremsstrahlung shape [11] or reference similar to that described in [7], although it would be best to generate using X-ray source response described in [8]. The basic KED analysis [1] performs a log-log transformation prior to fitting the energy regions. The assumption is that the log-log is generally insensitive to small changes in the overall bremsstrahlung shape. What's important is that the simulated reference and solution assay utilize the same bremsstrahlung shape.

4. Proposed Simulation of HKED Spectra

We have proposed, and begun developing the requirements specifications, for a new HKED software application, called SIMHKED, that addresses all the major needs outlined in an earlier section. The basic program flow of SIMHKED is shown in Figure 2 where three main program flow branches are depicted: (1) creation and jugging of input and product solutions; (2) X-ray tube and HKED instrument geometries; and (3) HRGS instrument geometry. The results of the 3 branches are fed into a computation section where the desired KED, K-XRF and HRGS spectral data are generated. The resultant spectra can then be analyzed in the standard way [1] for training, and other pedagogical reasons, producing U, Pu, U:Pu with, and without, Minor Actinide (MA) concentrations. In addition there is the option to generate spectra from the FP purified waste. Alternatively, the spectral data can be consumed for the purpose of developing a second generation of analysis algorithms.

The first branch addresses the need for creating virtual input and product solution samples for a few select fuel assemblies, for any level of BU and cooling time, and for any method of separation, *e.g.*, PUREX or pyrochemical. Included in the process are delays after jugging to accommodate any simulations necessary for hydrolysis effects and evaporation correction.

The second branch implements the modeling of the HKED instrument geometry for both the SSV and CSV arrangements. The requirement specifications stipulate that the software possess a simple geometry composer so as to allow the flexibility of easily making small changes, visually, in the HKED geometry including the sample changer. This will facilitate the investigation of sources of error such as sample positioning and variations of the X-ray tube output (HV, current and target degradation).

The last branch addresses the option of including HRGS results which requires the HRGS instrument geometry. In the SSV HKED instrument geometry it is assumed that the HRGS views the fission product spectrum in the passive mode whilst in the CSV geometry the mode may either be passive or active. Similar to the HKED instrument geometry, there will be a simple geometry composer that allows simple modification of the HRGS instrument geometry.

The last two branches require the detector efficiencies which can either be modeled or have the flexibility of being obtained externally via measurement or some other means, *e.g.*, ISOCS.

The requirements specifications indicate that there needs to be the flexibility of minimizing processing time, sacrificing accuracy, to accommodate situations where rapid spectral development is required but accuracy is not.

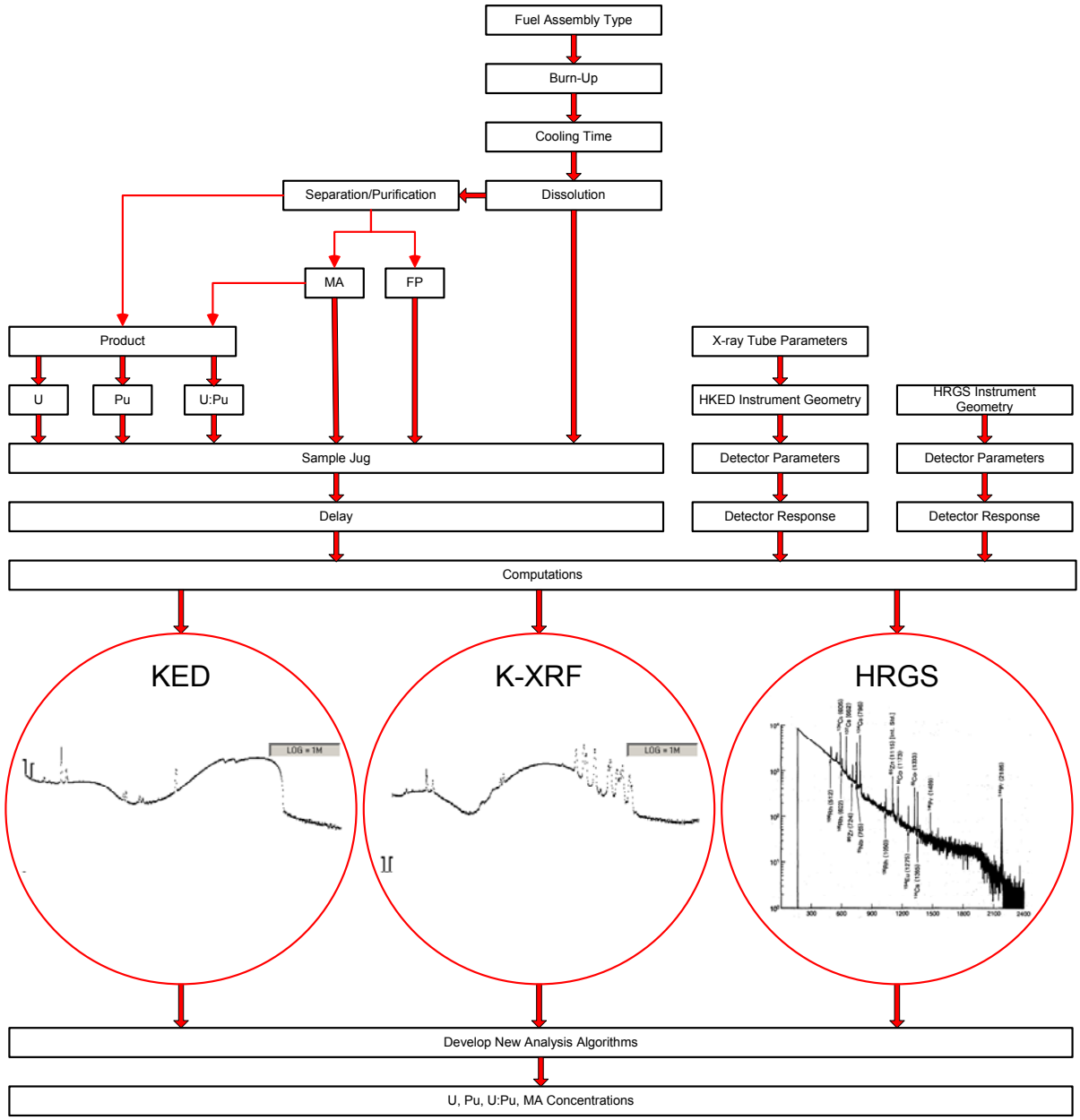

Figure 2 Proposed program flow for SIMHKED.

5. Discussion

The main goal of this paper is to capture the requirement specifications for the simulation of HKED and HRGS spectral utilizing what has been previously developed from the literature. The previously developed methods [7,8,9] are quite satisfactory at generating K-XRF spectral data however there are no provisions for BU, spent fuel cooling, FP excitation, hydrolysis, X-ray tube and sample position effects as well as the generation of KED and HRGS spectral data. This will all have to be developed.

SIMXRF requires measured reference file and calibration but does not require any MCNP modeling. Just photo-atomic data and is very fast (seconds) in generating simulated data. The Berlizov approach is rather time consuming but does not require any prior reference spectrum or calibration data. The Berlizov method does have the limitation with the limited Kβ's. The NCSU approach seems plausible and similar to the Berlizov work, however, there was no detail provided on how the X-ray source is modeled or the computation time impact of including coherent scattering and whether or not it is really necessary.

Berlizov indicates to save computation time the bremsstrahlung is only generated at and above the U, Pu KED regions. For proper analysis, including Th and dead-time considerations, the bremsstrahlung source energies must be extended to much lower energies. It is unclear as to what increase in the computation time will be realized if the energy domain is extended.

Berlizov indicates that there are differences in the MCNP libraries that can lead to differences in absolute yields of K-XRF data and absolute, *i.e.*, standalone, concentration analysis. This needs to be investigated and compared to any related potential biases encountered by the work by NCSU.

The feature in MCNP that leads to averaging of Kβ lines needs to be addressed since standard algorithms use the UKβ1-UKβ3 lines under certain situations. Note that this is a problem with Berlizov's work, but apparently not an issue with the work reported by NCSU.

Berlizov's work does not include Minor Actinides (MA) although SIMXRF does. It is not clear if the NCSU work includes MA, however, it appears as though this functionality exists.

6. Conclusion

The primary goal of this paper is initiate the first phase of the requirements specifications for a general purpose HKED simulator. The purpose of generating simulated HKED data is for both pedagogical and algorithmic development purposes addressing the challenges and shortcomings of the analysis algorithms of today. An investigative search effort was conducted to compile a list of the known work related to the subject and a detailed technical comparison is provided.

7. Future Work

This paper is really the beginning of the beginning of generating the specific requirements for simulating HKED data. A more detailed study of the three existing [7,8,9] models is necessary and their relative strengths and weaknesses better understood. Once the study is complete then a prototype version of SIMHKED will be developed.

Efforts will be made to secure measured data and associated HKED instrument geometries to assist in the development of the optimum model for confirmation purposes.

As the program develops it may become necessary to include other sources of excitation other than the photon flux from X-ray tubes and to extend the models to include the scanning of MOX fuel rods.

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Measurement of delayed neutrons from ^{235}U produced by cold neutron interrogation

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Abstract

The measurement of delayed neutrons for demonstrating the presence of fissile material is a well established method. However, the use of cold neutrons as an interrogating agent opens up wider perspectives. We exploited the combination of the high intensity of the neutron beam and the high cross section of the cold neutrons provided by the Budapest Neutron Centre.

The distribution of the half lives of the delayed neutrons shows continuous character, it used to be classified into 6 groups with half lives from 0.18 s to 55 s. From this distribution we investigated in two time windows, a short one up to 40 ms using a chopper rotating at 275 rpm, and a long one up to 8 seconds using a programmable beam shutter.

At 275 rpm the linearity between the integral of the counts and the ^{235}U content between 100 and 800 µg has been demonstrated. Similar proportionality has been observed in the case of beam shutter measurements above 400 µg ^{235}U content. We can conlude that this method is a sensitive one for detecting small amounts of ^{235}U and is worth for further development.

Keywords: delayed neutrons; cold neutron beam; NDA; U content

Taking Pu inventory of Pu-Be neutron sources by NDA methods in Hungary

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Abstract:

The presence of Pu-Be neutron sources in Hungary is an issue both for safeguards and security. About 200 sealed Pu-Be sources were imported to Hungary in the period 1960–1985. Being left from industrial applications, most of them are out of use recently. The Pu content of these sources was not declared upon delivery, and it remained basically unknown (and this is the case in several countries as well). Neutron output only was provided by the supplier. The missing information is relevant for safeguards, nuclear safety, physical protection, illicit trafficking, and material management purposes. The State and facility inventories were, and still partly are, based on rough estimated values. The neutron output of the sources ranges from 10^4 to 10^7 neutron/s. Bookkeeping was based on a calculation of the domestic authority relying on the neutron output, using a specific yield value $6.17 \cdot 10^4$ n/s g Pu, assuming pure ^{239}Pu content. Estimated on this basis, Pu quantities in individual sources amounted to 0.1 – 178 g (nominal values). However, the neutron output depends very much on the actual isotopic composition. Since the sources contain also other Pu (and Am) isotopes with much higher specific activities, these data can be considered as an upper limit only.

Plutonium content of the sources was determined in the frame of the Hungarian support programme to IAEA safeguards by applying NDA methods. Gamma spectrometry and neutron gross and coincidence counting were used either as combined or standalone methods as well. Calorimetry served for calibration. The total Pu amount of 121 sources investigated so far proved to be a rough 16% of the sum of nominal values. Pu inventory of the rest is planned to be taken as well. Our method is offered for routine use also to other countries facing similar problems.

Keywords: neutron sources; safeguards, illicit trafficking; NDA; Pu content

1. Introduction

Verification of sealed PuBe neutron sources present in Hungary is an issue for safeguards and security. About 200 sealed Pu-Be sources were imported to Hungary during the period 1960–1985. Being left from industrial applications, a lot of them are out of use recently, and were temporarily stored at the Institute of Isotopes. The Pu content of these sources was not declared upon delivery, and it remained basically unknown (and this is the case in several countries as well). Neutron output and activity (calculated from the neutron output) were only given. The missing information is relevant for safeguards, nuclear safety, physical protection, illicit trafficking, and material management purposes. The State and facility inventories were, and partly are still based on rough estimated values. The neutron output of the sources ranges from 10^4 to 10^7 neutron/s. The bookkeeping was based on a calculation from the neutron output, using a specific yield value $6.17 \cdot 10^4$ n/s g Pu informally given by the manufacturer, assuming pure ^{239}Pu content. Estimated on this basis, Pu quantities in individual sources stored at the Institute amounted from 0.1 to 178 g. These figures entered into reports as nominal mass values. However, the neutron output depends very much on the actual isotopic

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composition. Since the sources contain also other Pu (and Am) isotopes with much higher specific activities, these data can be considered as an upper limit only.

In order to solve the long-lasting problem, a project has been started at the Institute for determination of the plutonium content in Pu-Be neutron sources applying NDA methods such as γ spectrometry and neutron counting [1,2]. The novel methods being developed apply high resolution γ-spectrometry (HRGS) and neutron (gross and coincidence) measurements, using a calibration taken from a series of measurements by a calorimeter (provided by JRC IPSC Ispra) [3]. These methods were used for determining the actinide mass of other, Cf-, Cm-, Am-containing neutron sources as well [4,5].

Correlation was found between the Pu content and the ratio *R/T* of real coincidence (*R*) to total (*T*) count rates [6,7]. In addition, HRGS alone proved to be a tool for verification of the Pu content [8]. Identification of unknown (smuggled and seized, accidentally found, or not documented) neutron sources is also possible by gamma spectrometry [4 - 6] or neutron coincidence counting [9,10] alike, or rather by both [11,12].

By using these methods, the isotopic composition and Pu content of 76 sources stored at the Institute were determined at first. In this way, Pu amount to be accounted for reduced substantially. Nominal Pu mass of some sources was overestimated even by more than an order of magnitude in some cases (where ^{239}Pu abundance is below 75 % of the total Pu content). The total amount of Pu of all the 76 sources stored in the institute was 563±15 g, in contrast with 2050 g according to the sum of declared values.

After the measurements, the sources were re-encapsulated in stainless steel holders to exclude Pu leakage into the environment (air), and were placed in special containers designed for transport and storage of neutron sources and nuclear materials. These containers, being accessible to IAEA and EURATOM inspections, are disposed in the country's preliminary store of nuclear materials out of use.

Extending to other sources being in the country, altogether 121 PuBe sources were assayed so far, whose total Pu content proved to be a rough 16% of the sum of nominal values. Pu inventory of the rest of Pu-Be sources being in the country is planned to be taken as well. A help for doing the same is offered also to other facilities/countries, facing similar problems.

The methods could also be used for determining the unknown Pu content of smuggled and seized sources.

References to other works in the literature are to be found in references of the papers [1-15].

2. Determination of the Pu content

Three methods have been developed at the Institute of Isotopes for determination of the Pu content of PuBe sources:

The neutron-gamma combination method requires the knowledge of the isotopic composition (isotopics) and the neutron output. Isotopics is determined by HRGS. The neutron output can be measured by a neutron detector or can also be taken from the certificate of the source [6]. If such a documentation is not available, or it is to be verified (e.g. for safeguards purposes), the gross (total) neutron output (singles) is to be measured. (Using the certified output, by counting singles, the efficiency of the neutron detector can be obtained). Besides, real coincidences (*R,* doubles) are also counted.

The Pu content can also be determined by using HRGS alone, without neutron counting [8].

And finally, by counting neutrons only, both totals *T* and real coincidences *R* (singles and doubles), determination of the Pu content is possible as well, without applying gamma spectrometry [7,9].

Upon measurement, the PuBe source is to be taken out of its container by long-shafted pincers and to be positioned for measurement, while monitoring the gamma and neutron dose rates continuously, then staying behind shielding.

2.1. Neutron measurements

The PuBe source taken out of its container is to be inserted to the measurement cavity of a coincidence neutron collar. Sources of low neutron output can be measured with commercial neutron coincidence counters.

However, the neutron output of PuBe sources can be as high as 10^7 n/s. When measuring sources of such a neutron yield with commercial coincidence counters, the coincidence dead time may be too high. Therefore a passive neutron coincidence counter with low detection efficiency was designed. It consisted of 9 - 14 ^{3}He counters embedded in a polyethylene moderator surrounding the sources to be measured [1,2]. The thickness of the moderator arrangement was chosen so that the detection efficiency is limited up to between 5 to 12 %, so that the coincidence dead time is not too high, taking into account the expected counting rates from sources of 10^4-10^7 n/s output [6,7]. On the other hand, the moderator thickness should not decrease too low, not to result in too low detection efficiency and coincidence rate.

A 9-14 channel signal processing unit, consisting of preamplifier-amplifier-discriminator chains, was built and connected to the counting tubes. The independent channels have the advantage of diminishing the dead time of signal processing. A commercial shift register or a list-mode pulse train recorder [13-15] can be used for coincidence counting.

Total (*T)* and real coincidence count rates (*R*) are to be recorded. Measured *R* and *T* are to be corrected for the respective dead times δ_t and δ_c.

2.2. Determination of the isotopic composition and age by HRGS

2.2.1. Spectrum evaluation by applying the computer code MGA^{++}

Determination of isotopic composition is based on measuring photopeak area ratios by applying relative efficiency (“intrinsic”) calibration, using the gamma peaks in the energy region of 59-600 keV, while the age is estimated at the same time automatically on the basis of the parent/daughter relation of ^{241}Pu/^{241}Am. (The age value can be compared with the production date from certificate).

The sources were measured by a large area (e.g. diameter 50 by 20 mm thickness, resolution 688 eV FWHM at 122 keV) planar Ge detector in a far field geometry. HRGS spectra were taken for 10–50 min counting time at 10-300 cm source-to-detector distance. The higher the neutron yield, the longer was the distance. The multi-channel analyzer (MCA) must be set up to 8192 channels to get a channel width of 0.075 keV, as required for applying the advanced Multi-Group Analysis computer code MGA^{++}. Isotopic composition varied in broad ranges. Relative ^{239}Pu abundances were determined as between 55 and 96%.

A typical Pu-Be spectrum is seen in Fig.1, indicating the principal gamma lines of ^{239}Pu [8].

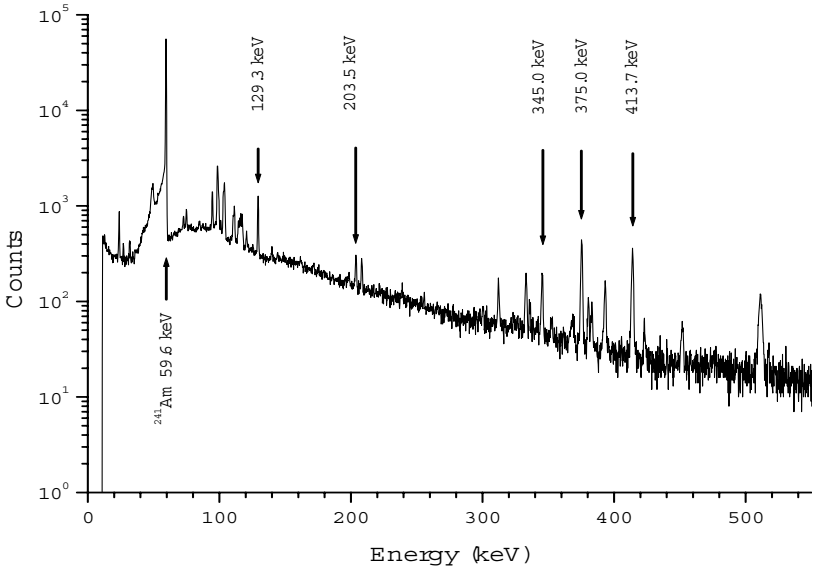

Fig.1. A typical 0-600 keV gamma spectrum of PuBe taken by a planar Ge detector [8]

2.2.2. Manual spectrum evaluation

The peak ratio technique is applied here as well, by using the intrinsic calibration method. Spectrum evaluation is done by the computer code GAMMAVISION and the MS-Excel software. By surveying the spectrum in the region of 94-106 keV complex X-ray range, the peak area at the 153 keV line of ^{238}Pu, at the 129 and 203 keV lines of ^{239}Pu, at the 160 keV line of ^{240}Pu, at 148 keV line of ^{241}Pu, at 208 keV line of ^{241}Pu/^{237}U (Am), and at the 59 keV line of ^{241}Am (^{237}U), one can decide whether the acquisition time is sufficient.

Spectra of a Pu-Be and an Am-Be neutron source, a pure Pu metal, and the background taken by a 150 cm^3 coaxial HPGe detector are shown in Fig.2. The Compton tail of the 4438 keV line of Be-containing sources and the effect of (n,γ) reaction increase the background. Because of the high background from PuBe sources of ^{239}Pu abundance of 70% or less, the MGA^{++} code becomes unusable.

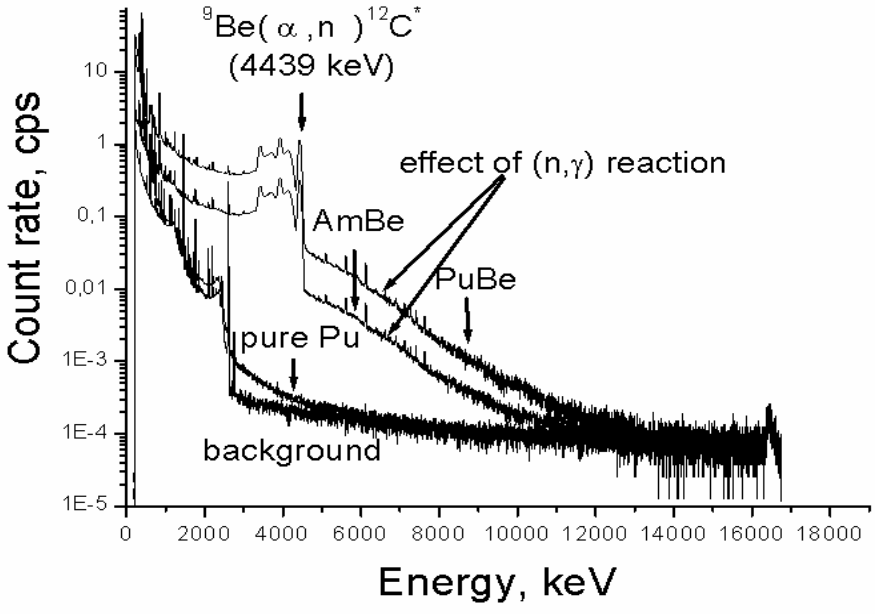

Fig.2. Gamma spectra of Pu-Be and Am-Be neutron sources, of pure Pu metal, and the background taken by a 150 cm^3 coaxial Ge detector

2.2.3. Correlation among Pu isotopic abundances

The isotopic composition of 8 Pu-Be sources determined by gamma spectrometry is plotted versus the ^{239}Pu component, f_{239}, in Fig.3 [8,9]. A nearly linear dependence of the isotopic abundances f_i of ^{238}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, and ^{241}Am on the ^{239}Pu component, f_{239}, was found, i. e. $f_i=a_i-b_i f_{239}$ (now i runs over the Pu isotopes and Am, from 1 through 5).

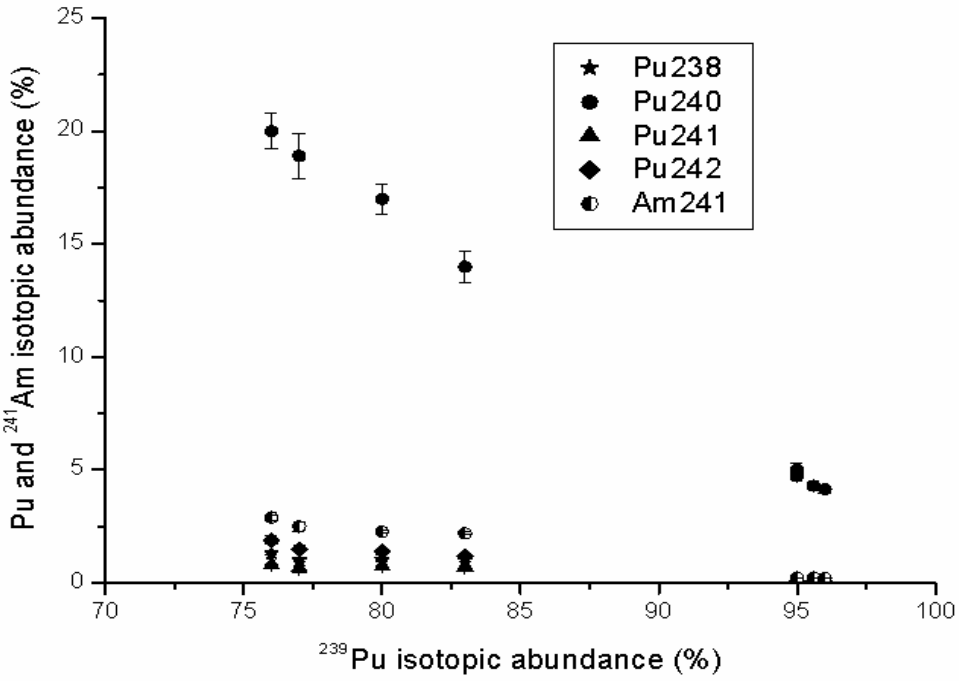

Fig.3. ^{238}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, and ^{241}Am abundances as a function of ^{239}Pu abundance in 8 PuBe sources, evaluated by MGA^{++} [8,10]

2.3. Combined neutron-gamma method

Gross neutron output of the source is to be measured for 2000–3000 s. The Pu content and the amount of the individual Pu isotopes is calculated from neutron output and isotopic composition, relying on specific (alpha,n) yields adopted for individual Pu isotopes and Am, according to the formula

$$m_{Pu} = \frac{N}{M \sum_i f_i y_i}, \qquad (1)$$

where N is the neutron output, M is the multiplication in the source due to secondary (neutron-induced) reactions (see below), f_i is the abundance of the i–th (Pu and Am) isotope (determined by HRGS), and y_i is the specific (alpha,n) yield of the i–th isotope. Summation goes over all the isotopes. Since f_i values are expressed in terms of percentage of the total Pu content, $\sum f_i$ exceeds 100% by the abundance of ^{241}Am, and the formula gives the Pu content only (considered to be a "true" value in contrast with the nominal one).

Specific (α,n) reaction yield (g_i) values for the Pu (and Am) isotopes were determined by starting with the specific alpha activities from the literature, multiplied by n/alpha ratios, which convert activity to neutron output. These ratios are sensitive to alpha energies, and thus are different for individual isotopes. The products obtained in this way are maximum attainable values. They can be less, if the Pu and Be constituents are incompletely dispersed and mixed in the source material upon production

of the sources. Thus, the products of the two factors are to be normalized. This was carried out by using the results of calorimetric measurements. The heat output was measured for 19 PuBe sources by the ANTECH Small Sample Calorimeter Model 601 provided by JRC IPSC, Ispra. The instrument was previously calibrated in the PERLA laboratory of the IPSC using certified reference materials. The sources measured by the calorimeter are used as secondary standards. By combining heat results with isotopics determined by gamma spectrometry, Pu masses were determined, relying on specific heat values from the literature. Using such a calibration, normalized specific (alpha,n) yields (g_i) were obtained, as follows:

^{238}Pu: 3.04E7 n/g-s
^{239}Pu: 8.84E4 n/g-s
^{240}Pu: 3.01E5 n/g-s
^{241}Am: 6.10E6 n/g-s.

The ^{241}Pu and ^{242}Pu specific yields, being of the order of 10^3, negligible at the usual isotopic ratios, were not considered. Similarly, the neutron yield from spontaneous fission of even mass number isotopes was neglected as well.

The multiplication in the source itself, (due to the induced fission in Pu and the (n,2n) reaction in Be), was taken into account by the correction factor *M*. This correction may amount to 25 – 30 % for the strongest sources, and can be determined by using coincidence measurements or Monte Carlo calculations. The results were practically the same by the two methods. An equivalent way is the use of the analytical formula [11,12]

$$M = 1+0.034\log m_{Pu} +0.0153\log^2 m_{Pu} +0.00283\log^3 m_{Pu} \quad (2)$$

and, if necessary, of iteration (usually 2 steps are sufficient).

The contribution of spontaneous fission neutrons to the coincidence rate is negligible. As compared to the effect of self-multiplication in Pu and the (n,2n) reaction in Be), it was estimated to be 1 - 3 % relative intensity for the smallest sources assayed. For more massive sources it is even smaller [2, 6].

Uncertainty of the Pu mass determined by calorimetry was mainly due to the error in determining the isotopics, which was in general taken to be 3 - 4 %, while the systematic error of neutron output was 10% at least. Nevertheless, the standard deviation among the measured sources of the same nominal (declared) neutron output was 2 -3 % only, therefore the precision of the combined method may well approach 5 – 6 %, even though the absolute value may differ from the real Pu mass by 10 – 15 %.

In addition to gross neutron counts, coincidence counts were also recorded. Real coincidences (after subtracting accidental coincidences) are due to secondary reactions, i.e. neutron-induced fission of the Pu isotopes (self-multiplication) and the ^{9}Be(n,2n)^{8}Be reaction. This multiplication in the source itself was taken into account by the correction factor *M*. This correction may amount to 25 – 30 % for the strongest sources, and can be determined by using simple arithmetical or Monte Carlo calculations. The results proved to be practically the same by the two methods.

2.4. Pure gamma spectrometry

Pu masses were also determined by pure HRGS, without neutron measurements. The method relies on absolute intensity measurements of the 129, 203, 345, 375, and 413 keV photopeaks of ^{239}Pu, applying attenuation correction, and taking into account the ^{239}Pu abundance f_{239}, determined also by γ-spectrometry from the same 0-600 keV gamma spectrum [8].

The source is taken out of its container and the 129, 203, 345, 375, and 413 keV photopeak areas of ^{239}Pu are measured by a large planar Ge detector for 10 – 50 min counting time in a far-field geometry (at 50 – 300 cm distance from the source, depending on its size and strength). Attenuation correction is applied, assuming a parallel beam falling on the detector surface and that the Pu-Be source has a cylindrical shape. For calculating self-attenuation in the source itself, the size of the PuBe filling inside source holders and the wall thickness were determined by X-ray radiography. The self-attenuation was calculated from the mass absorption coefficients of Pu and Be, with the assumption that the

source material was consisting of the intermetallic compound $PuBe_{13}$ of density of the literature value, 3.7g/cm^3. The actual density values were, however, determined from the calculated mass of the $PuBe_{13}$ compound (exploiting Pu mass values determined by calorimetry [3]), establishing the volume of the filling from the radiogram. Densities of 1.5 – 2.67g/cm^3 were found in the secondary standard sources, indicating that the material inside is a loose powder. At last, the unknown quantities, i.e. the actual density of a source and its actual Pu content, can be determined from the measured intensities of the photopeaks of ^{239}Pu, performing attenuation correction and calculation of the Pu content at the same time, by applying the MS-Excel far-field cylindrical calculation model.

The calculated count rates of the 5 principal ^{239}Pu gamma lines at 50 cm source-to-detector distance as a function of total Pu content (for the planar Ge detector mentioned above in Sect.2.2.1) are plotted in Fig.4 [8].

The advantage of the pure gamma method is the use of the 0-600 keV part of the spectrum only for estimation of both the isotopic composition and the Pu content, while the gamma line for Pu content evaluation is coming directly from ^{239}Pu, the main Pu isotope component of PuBe neutron sources. Uncertainty of the absolute value of the Pu mass is 10 – 15 %.

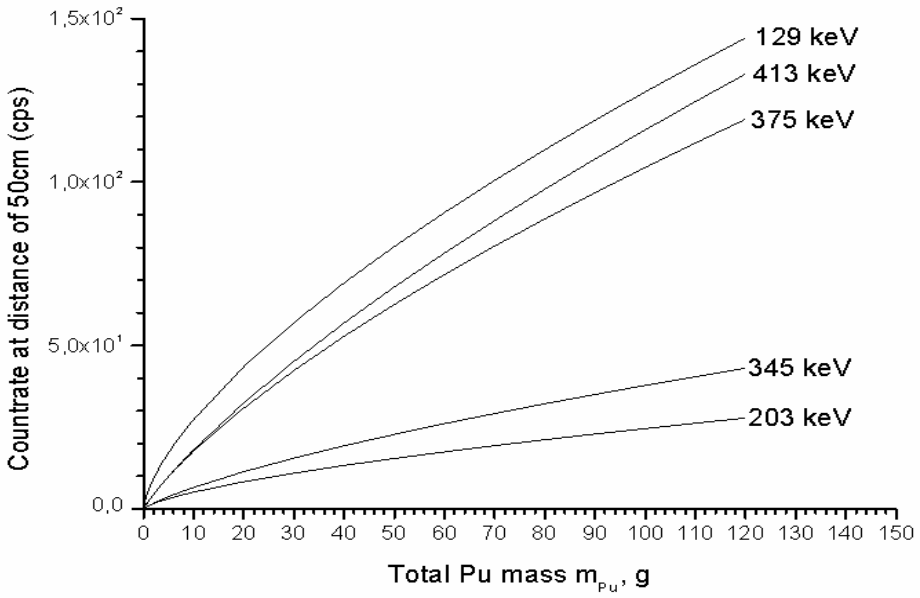

Fig.4. Calculated count rates of five gamma lines of ^{239}Pu at L=50 cm [8]

2.5. Pure neutron counting ("R/T method")

Pure neutron measurements are based on total (*T*) and coincidence (*R*) count rates, without γ-spectrometry. This so-called *R/T* method relies on a correlation established between the ratio *R/T* and the Pu content determined by calorimetry. Calibration of the *R/T* method for three neutron collars of 5, 11.3, and 28% efficiencies can be seen in the diagrams and in the corresponding formulae in Figs 5,6. Differences in ^{239}Pu abundances play a minor role in this method, as shown in Fig.6 [7].

The advantages of the pure neutron method are that there is no need of using a gamma detector and it is usable with Pu content of high burnup, where other methods cannot apply. However, analyzing the data measured by JCC-13 and JCC-31 equipments show that the *R/T* method needs a set of standard PuBe neutron sources. Since such sources were not available, 7 representative sources were selected from the 76 sources being stored at the institute, and their Pu content, having been determined by calorimetry, served as secondary standards. The main characteristics of these sources are shown in Table 1, indicating their Pu contents determined by various methods [11,12].

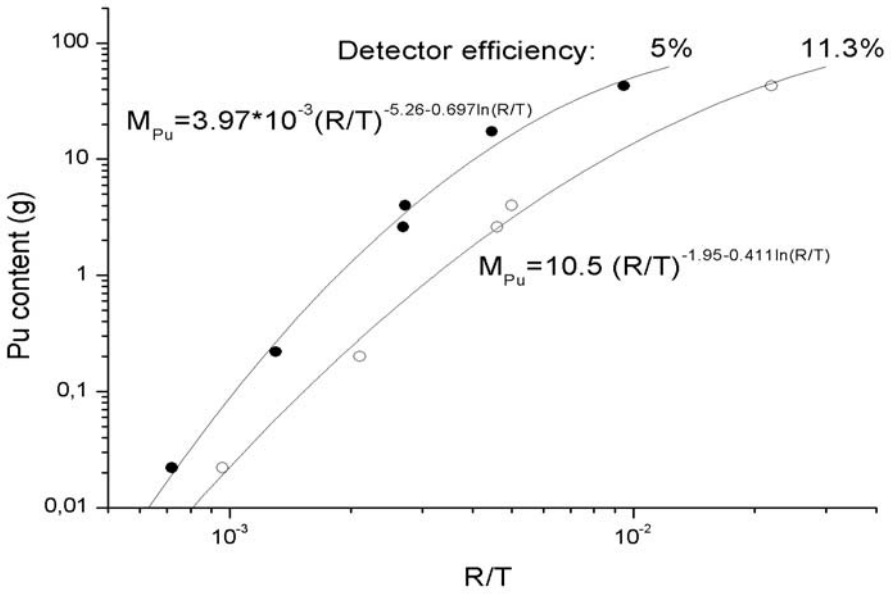

Fig. 5. Calibration of the *R/T* method against calorimetry [11,12]

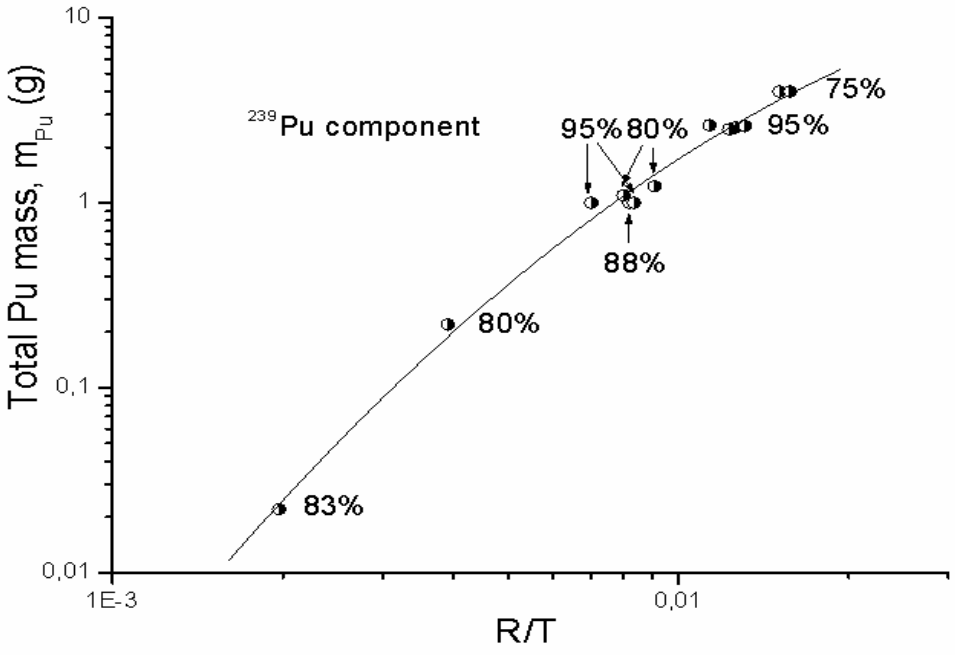

Fig.6. The relationship m_{Pu} - *R/T* by a detector of 28 % efficiency (JCC-13) [7]

The final result for the Pu content of a source can be given as the weighted average of the methods applied.

Declared neutron output, (n/s)	^{239}Pu fraction (%)	Pu content (g)				
		Nominal	Combined n-gamma	Pure gamma spectrometry	Calorimetry	Pure neutron counting
1.1×10^4	83.15(0.83)	0.18	0.022(0.003)	0.021(0.003)	0.0223(0.0041)	0.023(0.003)
1.1×10^5	79.76(0.80)	1.8	0.22(0.01)	0.225(0.004)	0.224(0.007)	0.23(0.03)
2.68×10^5	95.21(0.19)	4.0	2.48(0.13)	2.55(0.10)	2.62(0.08)	2.6(0.3)
2.26×10^6	76.92(0.82)	37	3.93(0.23)	4.0(0.3)	4.0(0.12)	4.4(0.7)
5.58×10^6	76.17(1.0)	45	9.62(0.63)	9.5(0.5)	10.4(0.31)	7.33(2.5)
1.1×10^7	75.7(0.91)	178	17.05(1.15)	17.8(1.6)	17.3(0.52)	15.3(2.9)
5.27×10^6	94.91(0.19)	85	42.1(3.0)	44.0(1.5)	44.3(1.3)	46.9(4.7)

Table 1: Main characteristics of 7 representative Pu-Be sources. Pu contents determined by various methods (errors are in brackets)

In order to find a detector-independent treatment, the normalized parameters $(R/T)_{norm}$ and $(T)_{norm}=N$ can be introduced. Various values of R/T from measurements of different detection efficiencies were normalized to 100% efficiency by the formula [7]

$$(R/T)_{norm} = (R/\varepsilon_2)/(T/\varepsilon_1) = \frac{R}{T}\,\varepsilon_1/\varepsilon_2, \qquad (3)$$

where ε_1 and ε_2 are instrumental constants depending on the parameters of the experimental setup. Here, $\varepsilon_1=\varepsilon$ and $\varepsilon_2=\varepsilon^2 exp(-P/\tau)[1-exp(-G/\tau)]$ approximately, where ε is the absolute detector efficiency, P is pre-delay, G is the coincidence gate length, and τ is detector die-away time.

Isotopic composition can be estimated by representing $(R/T)_{norm}$ as a function of $(T)_{norm}=N$, as is shown in Fig.7 [10].

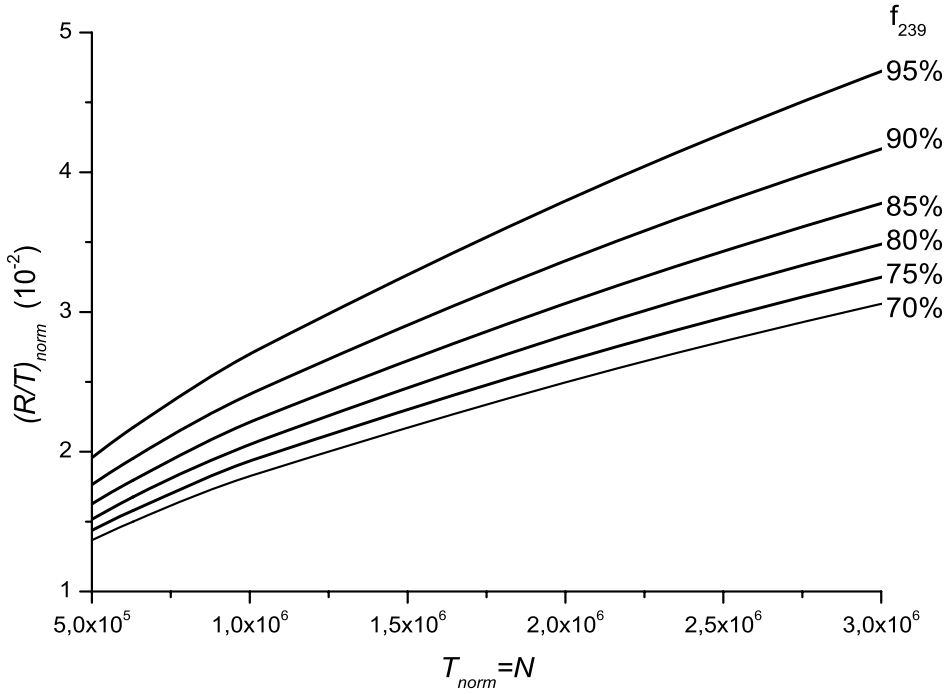

Fig.7. The normalized ratio $(R/T)_{norm}$ versus neutron output, $(T)_{norm}=N$, at various ^{239}Pu fractions [10]

3. Serial measurements, re-encapsulation, and disposal of PuBe sources stored at the Institute of Isotopes

In a comprehensive measurement programme, neutron output, isotopics, and Pu content of 76 Pu-Be sources, being stored in the Institute, were determined. As a result, it has been turned out that facility and State inventories are based on incorrect, highly overestimated values. The measurements resulted in a total Pu amount of about a quarter of the sum of the nominal values reported. A new Pu inventory was prepared and reported to the domestic and foreign (IAEA, EU) authorities.

70 measured Pu-Be sources were encapsulated. In the course of this, the Pu-Be sources in their present form – i.e. in their old capsules - were sealed into stainless steel (KO-36 type) holders with a wall thickness of 1.5–2.0 mm. The holders were sealed by using Argon gas-protected welding. The new holders (diam. 22.2–42, length 50–147 mm) contain 1–4 old Pu-Be sources in order to decrease the required storage room for the sources in the final disposal. After re-encapsulation, the batch numbers of the old sources (as used in safeguards accountancy) were engraved onto the surface of the new holders to help their future identification. These containers, being accessible to IAEA and EURATOM inspections, are disposed in the preliminary store of nuclear materials out of use.

The newly encapsulated sources were placed in special containers for shipping and storing according to the ISO 9001 quality assurance system. The containers were shipped to the radioactive and nuclear waste disposal facility in Hungary, where they remained accessible to identification and verification for authorities.

A series of further measurements was carried out in 2009-2010 for determining the Pu content of 45 sources. Among them 42 were of low mass (below 3 g each) of high burnup (50-70% ^{239}Pu) Pu content. Their measured Pu contents along with the respective data of the secondary standard sources are plotted in Fig.8 as a function of the parameter *R/T*.

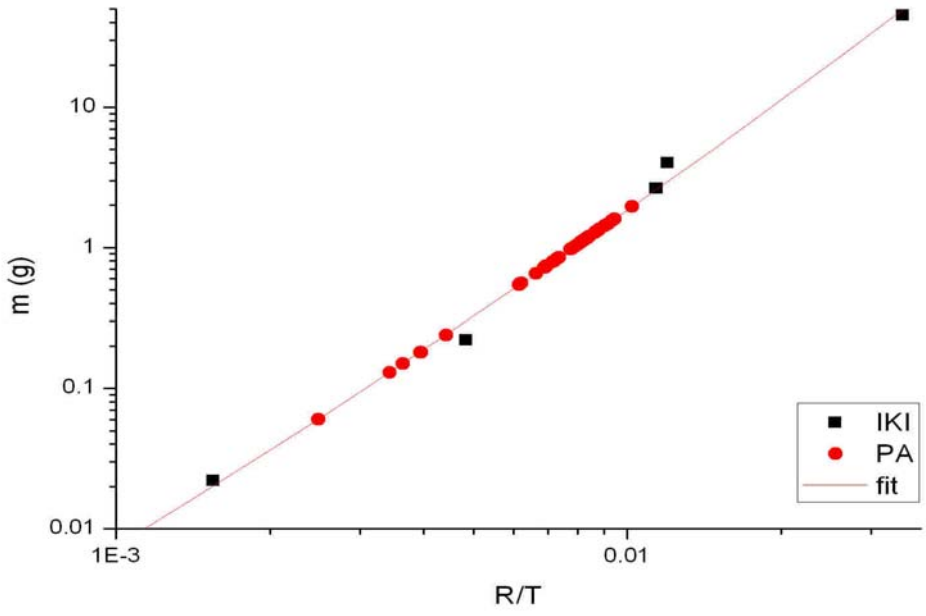

Fig.8. Results of measurements carried out recently: Pu content of a series of sources along with the data of the secondary standards plotted as a function of *R/T*.

The ratio *R/T* of the 45 sources is plotted in Fig. 9 as a function of *T*, at various ^{239}Pu fractions. Lines correspond to a given ^{239}Pu ratio of the sources of various neutron output.

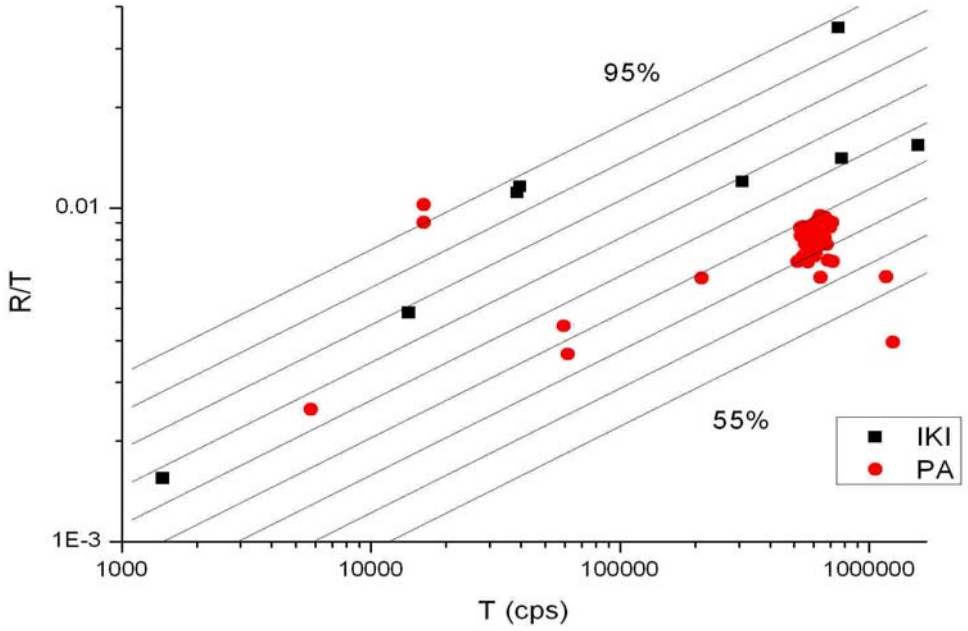

Fig.9. The ratio *R/T* as a function of *T* at various ^{239}Pu fractions

4. Summary

The Pu content of 121 Pu-Be sources measured altogether so far and corrected for safeguards accountancy: total 650.6 ±15 g of Pu, instead of 4043 g, the sum of nominal values.

5. Acknowledgements

IAEA is gratefully acknowledged for providing loan of the shift registers. IPSC Ispra is also thanked for making available the calorimeter and the neutron coincidence collar Type JCC-13 for test purposes. This work was supported by the Hungarian Atomic Energy Authority and Paks Nuclear Power Plant.

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An Estimate of Prompt Critical Mass of a Fissile Nuclide Including Capture within the Point Model

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Abstract:

A simple way to estimate the critical mass of a bare sphere of fissile material is developed without having to know the complexities of neutron transport theory. In addition to fission and scattering we include the parasitic capture cross section, capture acts as another neutron loss mechanism. Applied to smaller bodies the method can be used to calculate the fast fission multiplication factors needed for nondestructive assay of fissile materials by multiplicity counting. We illustrate the distinction between the leakage self multiplication and total multiplication and how the expression developed represents the distinction within the point model. We give examples of use benchmarking the expression to Monte Carlo data.

Keywords: multiplication, coincidence counting, multiplicity counting, point model, criticality

1. Introduction

In the field of nondestructive assay by correlated neutron counting the point model is widely used to quantitatively interpret the observed Singles, Doubles and Triples rate to extract unknowns, such as item mass and multiplication, of interest. The time independent, one energy group, point approximation for self-multiplication [1] is based on a neutron balance equation and provides a convenient parameterization of the item which is also pedagogically simple because the details of the neutron transport are hidden in a few interaction probabilities. In order to be able to estimate the properties of an item it is necessary to have a way to estimate the fate of the neutrons. Most often resort is made to Monte Carlo calculations. In this work we take an alternative look at the problem from an analytical perspective and show how the problem can be tackled in a straightforward way, at least for simple bodies, without needing to delve into the details of neutron transport theory or numerical simulation tools. This is useful from an educational perspective in the class room but also has practical applications when, for instance, we want to develop scaling (or interpolation) rules between reference and/or measurement items. To illustrate the method we calculate a rather extreme result, that of the prompt critical mass of a bare sphere of pure fissionable material. Typically assay items of interest in the safeguards domain are far from critical.

2. Method

Suppose we launch neutrons at a steady rate S per second into the item. Once dynamic equilibrium has been established the average rate at which neutrons leak out of the system, L, is given by the following simple balance equation:

$$L=S-F+v.F-C \qquad (1)$$

where F is the induced fission rate and C is the parasitic loss to capture and, for the present discussion, we shall neglect rare but exotic multiplying reactions such as (n,3n). Fission event removes the incident neutron but on the average liberates a total of v prompt neutrons per fission. Neutrons that leak are assumed never to return for the present discussion. That is the item is bare, or unreflected.

The rate at which neutrons emerge can be expressed in terms of the initiating source term and the leakage self-multiplication as follows:

$$L=S.M_L=S.(1+[(v-1).F-C]/S) \qquad (2)$$

or

$$M_L=1+[(v-1).F/(S+v.F) – C/(S+v.F)][1+v.F/S] \qquad (3)$$

Now we recognize (S+v.F) as the total rate at which neutrons are released into the system and each neutron present in the system at a given instant is seen to be just a trial for induced fission and capture – that is very neutron samples the possible fates. Thus we can define the average probability of fission, p_f, and the average probability of capture, p_c, as follows:

$$p_f = F/(S+v.F) \qquad (4)$$

$$p_c = C/(S+v.F) \qquad (5)$$

we note also that:

$$v.p_f=(v.F/S)/(1+v.F/S) \qquad (6)$$

from which we have the following relation

$$v.F/S = v.p_f/(1-v.p_f) \qquad (7)$$

Back substituting into equation (3) and re-arranging we obtain the following expression for the leakage self-multiplication in terms of the fission and capture probabilities and the mean number of neutrons emitted per fission:

$$M_L = (1-p_f-p_c)/(1-v.p_f) \qquad (8)$$

Because every neutron must either escape, induce fission or be lost to capture by the law of normalized probabilities we may write:

$$p_L+p_f+p_f=1 \text{ or } p_L= (1-p_f-p_c) \qquad (9)$$

which allows us to write equation (8) for the leakage self-multiplication factor in terms of the product of the total self-multiplication factor M_T and the probability of leaking from the system in the following fashion:

$M_L = p_L.M_T$ (10)

where

$M_T = 1/(1-v.p_f) = 1/(1-k)$ (11)

and we have introduced $k= v.p_f$ the usual prompt neutron multiplication factor familiar from simple reactor theory and which is equal to the number of neutrons in a generation divided by the number in the preceding generation.

Now, for an external detector p_L and the detection efficiency per neutron are seen to be naturally coupled as a product. Note too that from our neutron balance equation we can also be used to partition the events, that is we have:

$F = p_f.S.M_T$ (12)

$C = p_c.S.M_T$ (13)

$L = S.M_L = p_L.S.M_T = (1-p_f-p_c).S.M_T$ (14)

A crucial observation is that the probabilities of fission p_f and of capture p_c are simply related through the corresponding macroscopic capture fission and capture cross sections as these dictate the relative fate the between the possible next interactions. We can express this algebraically by:

$p_c = (\Sigma_c/\Sigma_f).p_f$ (15)

From (9) and (15) it is now clear that if we can estimate p_L by some means then knowing (Σ_c/Σ_f) for a given item (material and geometry) we can also obtain p_f and p_c and given v, the mean number of prompt neutron emitted per induced fission, the solution to the question of what values the multiplication takes immediately follows.

Inspection of (11) and (12) reveals that as $k=v.p_f$ approaches unity the fission rate increases without limit. That is to say a single neutron can result in an infinite number of subsequent neutron generations and we have the possibility of a prompt neutron run-away chain reaction (one where delayed neutrons are not needed to sustain the fission chains). Thus we see that:

$k=v.p_f = 1$ (16)

is the condition for prompt neutron criticality.

We shall now turn our attention to estimating the escape probability, p_L. In the point model approximation the value of p_L (and also incidentally of p_f and p_c) is unique since we have no spatial or energy dependence to contend with. This is a major simplification which implies items must be small compared to the neutron mean free path, typically dry items in which only fast neutron interactions matter, and conform to the model in the sense that space and energy averaged parameters apply. However, we have to realize that in real life the escape probability depends on the size and shape of the item as well as on its density and composition. The dependence is also somewhat subtle because in addition to fission and capture the material can also scatter neutrons redirecting them and hence extending the distance they travel inside the body and therefore also increasing the chance of fission. Our simple neutron balance equation did not need to consider scattering since the neutron population (number) remains unaffected by scattering.

Let us define the total macroscopic interaction cross section Σ_t of the medium comprising the item as the sum of the three reactions channels of interest assuming the medium is uniform and homogeneous. That is:

$$\Sigma_t = \Sigma_f + \Sigma_s + \Sigma_c \qquad (17)$$

To proceed let us take, purely as an illustrative example, a uniform spherical body in free space as our item. Let the radius be R and define the parameter x as follows:

$$x = \Sigma_t.R \qquad (18)$$

If neutrons are uniformly distributed and moving isotropically in the item (which they will be for the point reactor approximation if not exactly so for large bodies) then an algebraic expression for the escape probability P_0 from a sphere without any interaction exists [2]:

$$P_0 = (3/8).(1/x^3).[(1+2.x).\exp(-2.x) + 2.x^2 - 1] \qquad (19)$$

It follows that $(1-P_0)$ is the probability that the neutron will suffer a collision of some kind and since the chance of this being a scattering event is just Σ_s/Σ_t we can write the probability, γ, that a neutron born in the body will scatter simply as:

$$\gamma = (\Sigma_s/\Sigma_t).(1-P_0) \qquad (20)$$

We must now estimate the importance of multiple scattering on the fate of the neutron but we shall do so only in a crude way to indicate the way for a more refined treatment. Imagine that each scatter simply redirects the neutron so that it behaves like a new source particle and so begins a fresh attempt at journeying to the outside world. This is not strictly true of course since we expect the spatial distribution of events to be different for the scattered neutrons compared to source events because in a scattering medium they will have a tendency to diffuse away from their point of origin and the neutron density will tend to an eigenvalue flux shape [3]. However, in the point model we do not have any spatial dimensions so the discussion is strictly moot. Exactly the same situation applies to the enumeration of p_f in that the spatial distribution of induced fissions will not be uniform throughout the sphere and hence our use of (19) can only be approximate in any case – most suitable for small bodies which can be treated in the one collision approximation.

From the multiplicative property of combining probabilities for independent events we find that the probability of a number, call it *n*, successive scattering events followed by neutron emerging from the body is:

$$P_n = \gamma^n.P_0 \qquad (21)$$

The probability that a neutron will emerge is therefore the sum that it will emerge without colliding plus the probability that it will emerge after 1, 2, 3 … etc. scatters. Within the approximate nature of our discussion we elect here to take *n* all the way to an infinite number of contributions and form the limit. Algebraically this is equivalent to summing a geometrical series in γ scaled by P_0 and so we have:

$$p_L = P_0/(1-\gamma) \qquad (22)$$

Reed [3] remarks that as the number of scatters increases the “neutron gas cloud” inside the body will expand as if undergoing a random walk so that a large number of collisions is not to be expected before the typical neutron escapes from a compact item. Therefore, in some sense, a bounding number of

collisions is expected to exist which means *n* should be kept finite. But here we shall let the problem quench itself rather than worry about the details of what amounts to the spatial distribution.

Recall our condition for prompt criticality is:

$$p_f = (1-p_L)/(1+\Sigma_c/\Sigma_f) = 1/v \qquad (23)$$

which upon substituting from (22) into (23) becomes:

$$p_L = P_0/[1-(\Sigma_s/\Sigma_t).(1-P_0)] = 1 - [1+(\Sigma_c/\Sigma_f)]/v \qquad (24)$$

To get a feel for this, if v=3 and capture is negligible equation (24) tells us that at prompt criticality p_L=2/3, meaning most neutrons escape! So we can see the benefit to reflecting neutrons back into the body as a means to reduce the critical mass. The full solution for P_0 obtained from equation (24) and expressed purely in terms of nuclear properties of the material is as follows:

$$P_0 = \theta.[1 - (\Sigma_s/\Sigma_t)] / [1 - y.(\Sigma_s/\Sigma_t)] \qquad (25)$$

where

$$\theta = 1 - [1+(\Sigma_c/\Sigma_f)]/v \qquad (26)$$

With the numerical value of P_0 obtained from equation (25) we can now, with the help of equation (19), solve for the geometrical parameter *x* which in turn gives us the critical radius and thus from the material properties of the body under consideration the critical mass. The solution for *x* must be done numerically but this is a trivial challenge.

3. Illustration

To illustrate the approach just developed we estimate the prompt critical masses of pure ^{235}U and ^{239}Pu spheres. Microscopic fission, capture and scattering cross sections, σ_f, σ_c, and σ_s, respectively, averaged over the fission spectrum were taken from the KAERI website [4]. For the present work we included both the elastic and inelastic scattering contributions into σ_s neglecting energy loss. For ^{235}U we have σ_f=1.235b, σ_c=0.08907b and σ_s=(4.566+1.804)b. In the case of ^{239}Pu we have σ_f=1.800b, σ_c=0.05294b and σ_s=(4.394+1.460)b. The metallic densities of the pure isotopes (molar masses of 235.0439 g.mol^{-1} and 239.0522 g.mol^{-1}, respectively) in the normal state near to room temperature were taken to be 18.71g.cm^{-3} and 19.42g.cm^{-3} for ^{235}U and α-phase ^{239}Pu respectively.

For the present calculations we ignore the weak delayed neutron emission that follows fission and consider only the prompt fission neutrons emitted (for prompt critical estimate this is legitimate because the dynamics are fast compared to delayed neutron periods). Prompt neutron yield estimates are taken from [5]. Since the yield varies almost linearly with incident neutron energy and over the energy range where the bulk of the fission neutron energy spectrum resides (0.1 to 6 MeV, say) the fission cross sections for ^{235}U and ^{239}Pu don't vary dramatically, for simplicity we take v evaluated at 2 MeV to be representative of the fission spectrum weighted value. Hence for ^{235}U we have v≈2.637 and for ^{239}Pu we have v≈3.163 prompt neutrons emitted per induced fission.

Using this nuclear data we obtain estimates of the critical masses for these two materials of 72.4 and 12.1 kg for ^{235}U and ^{239}Pu respectively. We find that the prediction extremely sensitive to the input parameters and not surprisingly, given the assumptions of the model, the critical masses and dimensions obtained

are not exact. For example, depending on the density, the critical mass obtained by sophisticated neutron transport modeling for bare spheres of ^{235}U and ^{239}Pu are expected to be in the vicinity of 50kg and 10kg respectively [6]. These are less than our crude model but are of the correct order.

4. Discussion

The expressions for the leakage self-multiplication, M_L, and total self-multiplication, M_T, that appear in the point model expressions for Singles, Doubles and Triples counting rates are succinct and beguilingly simple but when one comes to apply them in practice one faces the problem of how to calculate the fission and capture probabilities p_f and p_c. In this work we have shown how p_f and p_c are related to each other and how they are also related to p_L, the leakage probability. The crucial step to the present analysis is the recognition that p_L can be approximated in terms of the uncollided escape probability P_0, if we accept some simplifying assumptions in the treatment of multiple scattering. Since the evaluation of P_0 is a simple geometrical problem for a uniform neutron population and in the case of a sphere has an analytical solution we can proceed immediately to an estimate of p_f and p_c based on the material and geometric properties of the item within the spirit of the point model and fast-neutron fission. This is a conceptually satisfying result for the physics student and brings an understanding of criticality into the realm of ready understanding without requiring high level neutron transport theory.

To illustrate the approach we chose to apply it to the rather extreme problem (certainly within the framework of the point model and routine non destructive assay within the civilian safeguards community) of the evaluation of critical masses. But it should be appreciated estimates for smaller items typically found in the fuel cycle, for instance fissile lumps present in waste, can also be calculated using the exact same approach. For small bodies (relative to mean free path lengths) we'd expect the agreement to be better and indeed we are limited to small bodies by the theory presented otherwise the spatial variation in the multiplication (i.e. p_f is not a constant) violates the point interpretational model for multiplicity counting and explicit steps need to be taken to handle this - by for example performing spatial integrals to obtain weighted powers of the multiplication [7].

In the educational setting the student may run examples to 'discover' salient behaviors for example how k and critical mass vary with material density and composition, such as for MOX powders rather than for metal. The treatment may also be extended in various ways. For example here we took the limit of $n \rightarrow \infty$ which would be most suitable if $\gamma << 1$, but small n-values can be studied as another bounding case. Within the undergraduate student's scope is an appreciation of the basic random walk problem which relates the size of the neutron cloud to n (and elementary diffusion theory) and hence allows a choice for the number of scatters before the neutron will have migrated out of the item. The choice of n does indeed exert an influence on the critical mass! For small bodies we may go to the other extreme and adopt the one scatter correction, however. Other shapes such as cuboids and ellipsoids can be studied analytically and the general method of Dirac chords can be introduced for general shapes.

From a practical standpoint we note that, based on detailed Monte Carlo calculations (which we shall not discuss here) that for a pure ^{239}Pu sphere k varies almost linearly with $r.(\rho/\rho_o)$, where r is the radius, ρ is the density (e.g. 16-20g.cm^{-3}, say, to cover room temperature forms) and ρ_o is the reference density used in the calculations and k vs $r.(\rho/\rho_o)$ fitting (for example 16.6g.cm^{-3}). In fact a cubic polynomial through the origin covers the entire sub-critical range rather nicely with the higher order terms introducing some slight sub-linearity. For $k<0.5$ (i.e. $M_T<2$) the simple linear approximation ($k \sim 0.18.r$, r in cm for 16.6 g.cm^{-3}

^{239}Pu metal) is rather good and under the condition that v remains constant this implies p_f scales in direct proportion to the radius. As a corollary according to the theory presented p_c and p_L also have correspondingly simple size dependence. This is a particularly useful result when extended to materials of interest in scrap and waste assay, for example ceramic fuel pellet fragments, where bounding limits must be placed on multiplication enhancement to the correlated neutron count rate, and it is the regime where our simple point model is able to provide justifiable quantitative guidance for practical measurements [8]. In additional to the instructional and education purpose behind this work our interest was very much engaged by these practical applications – for example quickly figuring out isotopic sensitivity of multiplication between pellets and defining a $^{239}Pu_{eff}$ mass in this context.

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On the Functional Form used to Represent the Response of Nuclear Calorimeters

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Abstract:

The current approach advocated in ASTM C1458 for how to represent the calibration curve of nuclear calorimeters is reviewed. In doing so, we also discuss the associated uncertainty propagation on the heat assay. We suggest that the ASTM C1458 standard is unnecessarily restrictive. Simple mathematical manipulation under reasonable approximations, consistent with ASTM C1458, shows that the thermal power can be expressed more simply as a direct low order polynomial function of the observed calorimeter signal. The benefit of taking this route is that it is analogous to the way many other non-destructive assay instruments are handled and one can therefore bring to bear established and familiar fitting tools.

Keywords: calorimetry; calibration; nuclear safeguards; fitting

1. Introduction

The spontaneous decay of radioactive materials is an exothermic process. If the radiations given off are absorbed in the materials of the item, heating occurs. The measurement of the rate that heat is liberated provides a way to quantify the amount of nuclear material present, provided that the relative isotopic composition of the radionuclides present in the item is known and the specific powers for each nuclide are also known. Such measurements are the domain of nuclear calorimeters and have been applied for decades in the nuclear fuel cycle. An ASTM Standard Test Method [1] covers the application of calorimetry to the non-destructive assay of the special nuclear materials Pu + ^{241}Am and ^{3}T. The control and accountancy of these materials is vital to the safe operation of nuclear facilities and to the mission of international safeguards. In the context of non-destructive assay nuclear calorimetry is often highly accurate provided a few basic assumptions are valid – such as the absence of chemical reactions that release or absorb heat.

In ASTM C1458 a particular functional relationship is adopted for the calibration curve. Specifically, a parameter known as the (differential) sensitivity, *S*, is defined and the change in sensitivity with thermal power is described by a linear function in power. There are reasons why one may expect such a behavior. For example, the thermal resistance of the air gap between the item and the inner walls of the calorimeter measurement cell changes with temperature (an effect that can in principal be reduced if a matched reference item could be placed in the reference cell of a twin cell calorimeter).

However, the consequence of making this assumption is that the relationship between the observed response and the power level becomes prescriptive and somewhat complex. In this work we revisit this assumption to show that it is valid to derive the thermal power directly from the observed signal. The benefit of doing this is that it makes available all of the curve fitting and visual presentation tools familiar to other non-destructive assay methods. A relaxation to the ASTM C1458 Standard Test Method would also permit justifiable alternative approaches to be used to the benefit of the calorimetry community while still being compliant to the standard.

2. Review of ASTM C1458 Approach

Denoting the calorimeter response at equilibrium with the item present by, V, and the baseline with no item present by V_0, we define the sensitivity S by the relation:

$$S = \frac{V - V_0}{W} \tag{1}$$

where W is the thermal power of the item and as a practical matter we take the net signal $\Delta = (V - V_0) \geq 0$. During calibration, pairs of V and V_o values are usually determined for a set of items covering the power range to be assayed.

The change in sensitivity with thermal power is assumed to trend linearly so that we write:

$$S = S_0 + k \cdot W \tag{2}$$

where S_0 and k are constant calibration parameters determined from analysis of experimental data. In performing an assay we are interested in estimating the thermal power from the net response observed and so invoking Eq (1) and (2) we may write:

$$W = \frac{\Delta}{S} = \frac{\Delta}{S_0 + k \cdot W} \geq 0 \tag{3}$$

Re-arranging and solving the quadratic relation in W which Eq (3) represents, and further recognizing that $S_0^2 \gg 4k\Delta$ for all typical systems, we have in a numerically robust form:

$$W = \left(\frac{-S_0}{2k}\right) \pm \sqrt{\left(\frac{S_0}{2k}\right)^2 + \frac{\Delta}{k}} \tag{4}$$

For a physical solution $W \geq 0$ and so for $S_0 > 0$ there are three cases to consider:

$k > 0$: The '+' sign in Eq (4) applies
$k < 0$: The '-' sign in Eq (4) applies
$k = 0$: $W = \Delta / S_0$ by direct inspection of Eq (1)

In addition to finding the value of W we also need to propagate the uncertainty in W that arises through the uncertainty in Δ and the uncertainty structure in S_0 and k. This is not discussed in ASTM C1458. For completeness we therefore consider it here in Appendix 1.

However, the foregoing approach seems awkward because W is the quantity required and $(V - V_0) = \Delta$ is the observed variable. It would seem logical in some ways to parameterize S directly in terms of Δ directly rather than via W.

3. Proposed Alternative Form

In this section we will derive an alternative expression for the thermal power based on comparable approximations. From Eq (3) we have

$$W = \frac{\Delta}{S_0}\left(1 + \frac{k}{S_0}W\right)^{-1} \tag{5}$$

If the slope, k, is small, then $S \sim S_0$ across the full dynamic range of operation, as depicted in Figure 1. That is, we may take it that:

$$\left|\frac{k \cdot W}{S_0}\right| \ll 1 \tag{6}$$

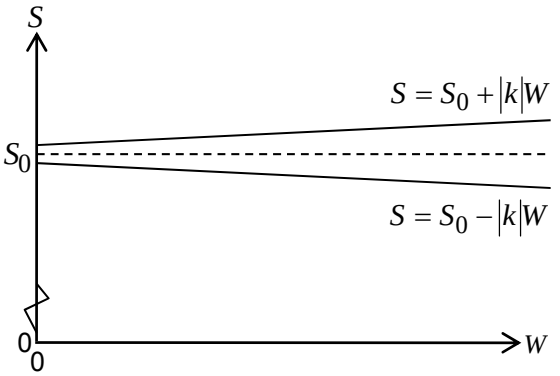

Figure 1: Assumed weak dependence of sensitivity S on the thermal power W in the alternative calorimeter response function.

Under these conditions we can use a first order expansion followed by substitution in Eq (5) to arrive at:

$$W \approx \frac{\Delta}{S_0}\left(1 - \frac{k \cdot W}{S_0}\right) \approx \frac{\Delta}{S_0}\left(1 - \frac{k\,\Delta}{S_0 S_0}\right) \tag{7}$$

or equivalently we may write

$$W \approx a\Delta + b\Delta^2 \tag{8}$$

with

$$a = 1/S_0 \tag{9}$$

$$b \approx \frac{-k}{S_0}\left(\frac{1}{S_0}\right)^2 \qquad (10)$$

Note that in Eq (10) the sign of k is implicitly defined coming directly from the behavior of S as a function of W. In the special case $k = 0$ which implies that $b \to 0$, we have no ambiguity to resolve. The solution is not hyper-sensitive to small ill-determined values of k. These are important secondary pragmatic advantages to the approach.

We emphasize that in deriving Eq (8) from Eq (3) and estimating values of a and b in terms of S_0 and k we have established a quantitative link between the two alternative approaches. However, in application of Eq (8) one does not need to work with S_0 and k at all. Rather the concept would be to treat 'a' and 'b' as empirical constants (model parameters) to be extracted directly from calibration data according to established best fitting tools and practices – the discussion of which is outside the scope of the present work.

We note that the same first order result (Eq (10)) may be obtained from the quadratic solution Eq (4) under the condition $\left|k \cdot W/S_0\right| \ll 1$ which implies

$$\frac{4k\Delta}{S_0^2} = \frac{4k\Delta}{S_0}\cdot\frac{1}{W}\cdot\frac{\Delta}{S_0}\cdot\frac{(S_0 + k\cdot W)}{(S_0 + k\cdot W)} = \frac{4k\Delta}{S_0}\cdot\left(1 + \frac{kW}{S_0}\right) \qquad (11)$$

So that with $\left|k \cdot W/S_0\right| \ll 1$

$$\frac{4k\Delta}{S_0^2} \approx \frac{4kW}{S_0} \ll 1 \qquad (12)$$

and we thus can expand the term under the radical of Eq (4) to first order accordingly.

Note that Eq (8) is simply a low order polynomial (in this case passing through the origin) and such forms are routinely used in the non-destructive assay community so that all of the familiar apparatus for dealing with related problems, and which are familiar in other ASTM standards & guides, can be adopted directly to the case of analyzing calorimetry data. For example, the DEMING curve fitting code [2], familiar to the safeguards community, allows W vs Δ fits to be performed on calibration data with uncertainties in each being propagated and fitting parameters along with the covariance matrix being generated according to the Deming definition of least squares but not carried out to full iterative convergence. In passing we note that inclusion of higher order terms in Δ could capture empirically in a straight forward way other functional dependencies of S on W other than the linear dependence assumed. Furthermore, if we have reason to believe V_0 can be treated as a constant, then W can be expressed as a second order in V (rather than Δ) but with the inclusion of a constant term also. For matched pair data each assay has its own unique baseline estimate and so this step would not be needed.

4. Application

As an example we consider a small diameter ARIES-I calorimeter calibrated at the Technical Area TA-35 facilities at Los Alamos. The power range is approximately 0.15 to 12 W. The data set is listed in Table 1.

The values of S_0 and k in the ASTM fit were estimated by minimizing the following sum formed over all 20 data entries:

$$\sum\left(\frac{S - [\![(S]\!]_0 + k.\Delta)}{S}\right)^2$$

Accordingly $S_0 \approx 20.0999$ and $k \approx -0.0141043$.

The values for a and b in the direct fit were obtained by minimizing the following sum formed over all 20 data entries:

$$\sum\left(\frac{W - (a.\Delta + b.\Delta^2)}{W}\right)^2$$

where W is the known power level (P in the Table to distinguish it from the unit) and Δ is the balance point condition. Accordingly $a \approx 0.0497449$ and $b \approx 1.79075x10^{-6}$.

For the present discussion the important point we wish to make is that the overall quality of the fit to the power, which we quantify by the percentage deviation, $100.\left(1 - {}^{W_{Fit}}/_{W}\right)$, is comparable for the two approaches. This is also shown in Table 1. We see that across the range the power values are replicated with almost the same quality for both methods.

Data Point	Thermal Power, P (W)	Net BP $\Delta=(V-V_o)$ (mV)	ASTM Fit $(1-W_{Fit}/W)$ (%)	Direct Fit $(1-W_{Fit}/W)$ (%)
1	0.149397	3.04779	-1.51	-1.49
2	0.149416	2.97206	1.03	1.04
3	0.149419	3.01622	-0.44	-0.43
4	0.149429	3.00531	-0.07	-0.06
5	0.149519	2.97505	1.00	1.01
6	0.266678	5.29968	1.11	1.12
7	0.266694	5.36324	-0.07	-0.06
8	0.266698	5.33884	0.39	0.40
9	0.266701	5.37620	-0.31	-0.30
10	0.266714	5.34980	0.19	0.20
11	0.266721	5.36328	-0.06	-0.05
12	0.266724	5.36050	-0.01	0.01
13	0.806535	16.28649	-0.52	-0.51
14	0.806552	16.24453	-0.26	-0.25
15	3.540575	71.17781	-0.27	-0.26
16	3.541870	71.18198	-0.24	-0.23
17	3.542023	71.28900	-0.38	-0.38
18	11.834910	235.71000	0.08	0.08
19	11.835420	236.14800	-0.10	-0.10
20	11.835674	235.21580	0.30	0.30

Table 1: ARIES-I calibration data

5. Conclusion

An alternative to the ASTM C1458 approach to treating the calibration function of a nuclear calorimeter has been derived. We contend that in some ways this is easier to handle and generalize. We have shown that it can work comparably well in practice. There seems to be no reason why the ASTM Standard Test Method should exclude such alternative approaches provided they are appropriately implemented and verified.

6. Future Work

Our intention in this article was to develop the mathematical justification behind admitting alternative data representation into the ASTM standard. We have illustrated the approach with a single data set. In a forthcoming article we intend to apply the method to a number of data sets acquired with a variety of calorimeters of different types.

7. References

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Appendix 1: Uncertainty Propagation for the ASTM C1458 Approach

We represent W by the function $W(S_0, k, \Delta)$ and by the usual uncertainty propagation rules note that the variance in W may be estimated to first order according to (in conventional and self explanatory notation):

$$\sigma_W^2 = \left(\frac{\partial W}{\partial S_0}\right)^2 \sigma_{S_0}^2 + \left(\frac{\partial W}{\partial k}\right)^2 \sigma_k^2 + 2\left(\frac{\partial W}{\partial S_0}\right)\left(\frac{\partial W}{\partial k}\right) \quad \text{(A1.1)}$$

where $\sigma_\Delta^2 = \sigma_V^2 + \sigma_{V_0}^2$ since $\Delta = (V - V_0)$ and V and V_0 are independent.

Note that in our expression for σ_W^2 we have kept the covariance term between S_0 and k since they are linked through the calibration while other cross terms are zero. To progress we must evaluate the partial differentials to be used in this expression.

From Eq (4) we have:

$$W = \left(\frac{-S_0}{2k}\right) \pm \sqrt{\left(\frac{S_0}{2k}\right)^2 + \frac{\Delta}{k}} \quad \text{(A1.2)}$$

from which we derive the following partial gradients:

$$\frac{\partial W}{\partial S_0} = \left(\frac{-S_0}{2k}\right) \pm \frac{1}{2k}\left[\left(\frac{S_0}{2k}\right)^2 + \frac{\Delta}{k}\right]^{-\frac{1}{2}} \cdot \left(\frac{S_0}{2k}\right) \quad \text{(A1.3)}$$

$$\frac{\partial W}{\partial k} = \left(\frac{-S_0}{2k}\right) \cdot \left(\frac{1}{k}\right) \pm \frac{1}{2k} \cdot \left[\left(\frac{S_0}{2k}\right)^2 + \frac{\Delta}{k}\right]^{-\frac{1}{2}} \cdot \left[2\left(\frac{S_0}{2k}\right.\right. \quad \text{(A1.4)}$$

$$\frac{\partial W}{\partial \Delta} = \pm \left[\left(\frac{S_0}{2k}\right)^2 + \frac{\Delta}{k}\right]^{-\frac{1}{2}} \quad \text{(A1.5)}$$

and it is understood that the choice of sign follows the decision path based on k in the text. In the special case $k = 0$, $W = \Delta/S_0$ and we have the following simplification:

$$\left(\frac{\sigma_W}{W}\right)^2 = \left(\frac{\sigma_\Delta}{\Delta}\right)^2 + \left(\frac{\sigma_{S_0}}{S_0}\right)^2 \quad \text{(A1.6)}$$

Appendix 2: Uncertainty Propagation for the Alternative Approach

We treat a and b as empirical parameters extracted from calibration as in Eq (8).

$$W = a\Delta + b\Delta^2 \quad \text{(A2.1)}$$

Forming partial differentials and propagating uncertainties in the usual fashion, we have

$$\frac{\partial W}{\partial a} = \Delta \quad \text{(A2.2)}$$

$$\frac{\partial W}{\partial b} = \Delta^2 \quad \text{(A2.3)}$$

$$\frac{\partial W}{\partial \Delta} = a + 2b\Delta \quad \text{(A2.4)}$$

$$\sigma_W^2 \approx \left(\frac{\partial W}{\partial a}\right)^2 \sigma_a^2 + \left(\frac{\partial W}{\partial b}\right)^2 \sigma_b^2 + 2\left(\frac{\partial W}{\partial a}\right)\left(\frac{\partial W}{\partial b}\right)c \quad \text{(A2.5)}$$

$$\sigma_W^2 \approx \Delta^2\sigma_a^2 + \Delta^4\sigma_b^2 + 2\Delta^3 covar(a, b) + (a + 2b \quad \text{(A2.6)}$$

with

$$\sigma_\Delta^2 \approx \sigma_{V_0}^2 + \sigma_V^2 \quad \text{(A2.7)}$$

since $\Delta = (V - V_0)$ and V and V_0 are independent observations.

Uncertainty Quantification in Spent Fuel Assay

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Abstract:

The Next Generation Safeguards Initiative is developing nondestructive assay (NDA) methods to detect diversion and to assay Pu mass in spent fuel assemblies. Uncertainty quantification is an important task in any assay method, and particularly for spent fuel assay. Typically, only some uncertainty components are straightforward to quantify. For example, assay repeatability is relatively simple to quantify. In NDA of spent fuel, a computer model (MCNPX for example) is used to predict the relation between three inputs: initial fuel enrichment (IE), fuel utilization (burnup, BU), and cooling time (CT), and multiple outputs. Example outputs include the detector response (DR) for any of various NDA measurement options such as differential die-away or passive neutron albedo reactivity and the effective fissile content or the content of particular fissile isotopes such as ^{239}Pu and ^{240}Pu. Computer model uncertainty is expected to be a large component of the total uncertainty in using measured IE, BU, and CT plus MCNPX and one or more measured DRs to predict fissile content and eventually total Pu mass. We can begin to quantify three error sources: (1) sample-specific departures from nominal assumptions, (2) measurement errors on the input IE, BU, and CT measurements, and (3) relying on interpolating MCNPX output at (IE, BU, CT) using an archived set of MCNPX outputs at specified values of (IE, BU, CT). For (1), sensitivity studies can assess the impact of modelling errors such as sample-specific departures from nominal geometries arising from small variations in how the spent assembly is aligned in the detector. For (2), Monte Carlo simulation using measured DRs to define importance weights or a numerical Bayesian strategy are recommended and illustrated. For (3), errors in code "emulators" that approximate the code output at untried input settings can be assessed using Gaussian process modelling for nonparametric fitting, or response surface estimation methods for parametric fitting.

Keywords: measurement error modelling, monte carlo, spent fuel assay, uncertainty quantification

1. Introduction

Estimating plutonium mass M_{Pu} in spent nuclear fuel assemblies (SFAs) using NDA is a challenge for the International Atomic Energy Agency (IAEA). After removal from reactors, SFAs are monitored for possible diversion, typically by cameras and visual inspections. In addition, gamma and neutron measurements are taken; however, inspectors usually check the consistency of the operator declarations for key parameters such as burnup (BU), initial enrichment (IE), and cooling time since removal from the reactor (CT). These radiation measurements and visual inspection do not attempt to estimate M_{Pu} in the SFA. Instead, they are intended to detect changes in the SFA that could be associated with Pu diversion.

Under the sponsorship of the Next Generation Safeguards Initiative (NGSI) an effort is underway to measure M_{Pu} in SFAs without such strong reliance on operator declarations of IE, BU, and CT, and without previous measurements of the SFA [1]. This effort is focused on non-destructive assay (NDA) techniques that can be used in a variety of surroundings (e.g., air, water, borated water), and that are

fast enough to not be a burden on the operating procedures at the nuclear reactor, spent fuel storage, or reprocessing facility. A few of these techniques will be integrated into one NDA system to allow a safeguards inspector to measure M_{Pu} in a SFA and to have a capability to determine whether spent fuel pins have been removed from the SFA.

The program aims to develop a combination of techniques, integrated in way to adequately estimate M_{Pu}. Fourteen NDA techniques are being investigated to varying degrees. No single technique can directly measure M_{Pu}, but each technique provides a measurement that contributes something toward estimating M_{Pu}. Some of the quantities that can be estimated from an individual measurement are: the average BU of an SFA; the ratio of the mass of the fissile isotopes to the mass of the fertile isotopes (fissile/fertile ratio); the ratio of the mass of elemental uranium to the mass of elemental plutonium (elemental U/Pu ratio); the mass of ^{235}U to the mass of ^{239}Pu ($^{235}U/^{239}Pu$ ratio); and a combination of the masses of the three fissile isotopes, ^{235}U, ^{239}Pu, and ^{241}Pu (fissile content) [2].

An MCNPX model has been developed for each of the 14 techniques. A library of SFAs has been created that contains a range of IE, BU, and CT values. The models are 17 fuel pins x 17 fuel pins, pressurized water reactor (PWR) assemblies with BU values of 15, 30, 45, or 60 GWd/t IE values of 2, 3, 4, or 5 wt % ^{235}U; and CT values of 1, 5, 20, or 80 yr [2,3,4]. The combination of 64 IE, BU, and CT parameters define the mass and spatial distribution of the many isotopes in the SFA and are representative of SFAs currently residing in nuclear reprocessing, storage, or reactor sites. The models are combined with the geometry of the detectors and a fixed source MCNPX calculation is performed with a variety of tally options to estimate the detector response (DR) of the detectors for each technique. The DRs can then be used to estimate the measured quantity of the technique. *And a major goal is to then use the measured quantity of the technique to assist in estimating* M_{Pu}. The MCNPX simulations can be performed with the assembly surrounded by either water, borated water (2200 ppm), or air. Each of the 14 NDA instruments has some DR that changes with some attribute of the assembly (e.g., fissile content, elemental U/Pu ratio, etc.).

At present, using the 64 IE, BU, and CT parameters as MCNPX inputs to define 64 idealized SFAs, the M_{Pu} *and the DRs for each of several individual techniques have been predicted for each assembly.* This paper focuses on one NDA technique, the passive neutron albedo reactivity (PNAR) method, so from here on, DR will refer to the detector response of PNAR and CR will denote its associated signature which is a ratio of count rates. As a step toward uncertainty quantification (UQ), we will model the measurement error processes associated with IE, BU, CT, and CR. For various measurement error variances, measured IE, BU, CT, and DR values will not lie exactly among the 64 modeled SFAs. To assess uncertainty, a brute force strategy is to re-run MCNPX at each hypothetical value of measured IE, BU, CT, and CR. However, because the MCNPX run times are long, the brute force approach of rerunning MCNPX for many slightly different IE, BU, CT, and CR values is not feasible. Therefore, for assemblies having parameters within the ranges of the 64 modeled SFAs, some type of interpolation will be used.

The following sections include a description of PNAR, measurement error modelling, MCNPX code output interpolation, example relative error standard deviation (RESD) results, discussion of "calibration" of a certain type, and summary.

2. PNAR

In the PNAR technique, neutron count rates of the SFA are measured both with and without a cadmium liner on the inside of the detector very close to the surface of the fuel. The spontaneous fission of ^{244}Cm in the SFA provides the source of interrogating neutrons. The Cd liner strongly absorbs thermal neutrons [5]. The PNAR signal – the so called Cd ratio, is the ratio of two measurements for which the only difference is the presence of the Cd liner in one case. The only difference between these two measurements is that in the "without Cd" measurement, thermal neutrons are reflected back into the fuel from the walls; therefore, the PNAR instrument can be thought of as interrogating the assembly with thermal neutrons coming from outside the assembly.

3. Measurement Error Modeling

The IE, BU, CT parameters and the CR will be measured with error [6,7,8]. One application for this effort is to reduce reliance on operator declarations of IE, BU, and CT. However, at this stage, we assume IE, BU, and CT are "measured," either by the operator or inspector. Although BU and IE are idealized concepts (thought of as an average IE or BU over the entire SFA), both can be defined and measured. And, both IE and BU must be "measured" indirectly using direct measurement of fuel parameters and a model relating observables to IE and BU. For example, a common way to "measure" BU measures ^{137}Cs because simple physics modeling suggests a linear relation between ^{137}Cs and BU (because a model is required, both IE and BU are indirectly measured, so for emphasis "measured" is in quotes).

Typically, metrology for nuclear safeguards adopts the notion of random and systematic error variances [9]. However, because we do not yet attempt to measure the total M_{Pu} in two of more SFAs, at this stage we restrict attention to random errors only, ignoring systematic errors. The simple error model for each measured quantity is therefore Measured = True x (1 + R), where $R \sim N(0,\sigma_{RSD})$ where $N(0,\sigma_{RSD})$ denotes the normal or Gaussian distribution, in this case with mean 0 and standard deviation, σ_{RSD}. For variance propagation, the normality assumption is not needed here but is made simply to be specific so that our results are easily duplicated by simulating normal random variables (rvs). We include a range of RESDs from 1% to 20%, and these values can be considered to result from adding systematic and random error variances although only random errors are currently used

4. MCNPX output interpolation

Figure 1 plots the M_{Pu} in arbitrary units (au) versus the CR for each of the 64 SFAs. As an aside, the concept of an effective Pu mass that accounts for the three main fissile isotopes (^{235}U, ^{239}Pu, and ^{241}Pu) is being pursued for this effort. However, in this paper, it is adequate to regard the data in Figure 1 as being elemental Pu mass. In Figure 1, the font size changes from small to large for CT = 1, 5, 20, and 80. The BU values are distinguished by color, and the integer 2, 3, 4, or 5 denotes the IE value. If CR is used alone to predict Pu, then qualitatively, Figure 1 indicates the resulting M_{Pu} estimate will have a very large RESD, too large to be useful. More quantitatively, Figure 2 plots the RESD in estimated M_{Pu} versus the CR. The average RESD from Figure 2 is also too large to be useful, but will serve as a basis of comparing different options for using CR.

The M_{Pu} was estimated either using a (a) nonparametric curve smoother, a (b) local linear smoother, or a (c) parametric fit. The fits from (b) and (c) are indicated in Figure 1, and the resulting RESDs were similar for all three. The RESDs in Figure 2 were computed using (c) and zero measurement error. Note from Figure 2 that across the 64 parameter values, the average RESD is approximately 14%. Although an RESD target value is not yet established, the target is probably 5% RESD or smaller. Therefore, efforts are ongoing to adjust for the rough ("jumpy") relation between M_{Pu} and DR as shown in Figure 1. The dominant effect causing the rough relation is the changing quantity of neutron absorbers, so techniques and measurements to adjust for neutron absorbers in the SFAs are being pursued. In this paper, we focus strictly on what can be learned from measurement error modelling combined with MCNPX modelling results such as those in Figure 1.

Using CR alone to estimate M_{Pu}, Figure 2 reports an average of RESD 14% (which does not include MCNPX model error or measurement error). Next, we consider two broad options to use CR to help reduce the RESD in the estimated M_{Pu}. First, use IE, BU, and CT to predict M_{Pu}, and then see whether adding CR as a fourth predictor reduces the RESD in M_{Pu} estimation. Second, and probably more appropriately, see whether using CR to modify the estimate of IE, BU, and CT prior to using IE, BU, and CT alone to predict M_{Pu} improves the RESD.

For the first option, we fit the modeled M_{Pu} as a function of IE, BU, and CT. Exploratory analyses such as the conditioning plot in Figure 3 suggest that linear, quadratic, and perhaps interaction terms should be included. Using `lm()` in R [10] to fit ordinary regression models [11], we omitted terms that did not significantly improve the fit as measured by the average squared error between the fit and M_{Pu} (as measured by the well-known F test [11]) The selected model is

$$\hat{M}_{Pu} = 2450.5 + 1311.0 \times IE - 26.5 \times BU - 17.3 \times CT - 23.1 \times IE.BU - 0.2 \times BU.CT + 133.7 \times IE^2 + 0.6 \times BU^2 + 0.2 \times CT^2 \quad (1)$$

where for example IE.BU is the product of IE and BU to capture the fact that the effect of BU depends somewhat on the value of IE, which is referred to an interaction term. Eq. (1) accounts for 99.7% of the variance of M_{Pu}, resulting in an RESD of approximately 2% in comparing M_{Pu} to $\hat{M}_{Pu}$. Although the fit of Eq. (1) is excellent, it does introduce mild interpolation error, so we report RESD results both (a) assuming Eq. (1) is the true model and (b) assuming the actual MCNPX model output M_{Pu} is the true model. In reality, neither is the true model because of modeling errors, but here we focus on error introduced by interpolating code output by comparing (a) and (b) in the next section.

Eq. (1) makes it simple to assess sensitivity of RESDs to σ_{RSD} for each of IE, BU, and CT. Simulation and an extension to Eq. (1) that includes CR as a fourth predictor (allowing for linear, quadratic and interaction effects as in Eq. (1)) indicate that the RESD is sensitive to IE whether CR is used or not. If CR is not used, then RESD is sensitive to BU. If CR is used, then the RESD is sensitive to both BU and CR. Qualitatively, CR provides an advantage in the sense that BU does not need to be well measured if CR is used. CT does not seem to be terribly important in either case (with or without CR as a fourth predictor)

Code emulation/approximation is increasingly used [12], particularly in situations where repeated code evaluation is required but too time consuming, such as in Markov chain Monte Carlo (MCMC) (next section) to estimate model parameters. Because of the low dimension (3 or 4 input parameters) and smooth relation shown in Figure 3, a parametric fit developed using exploratory data analysis is defensible. In other situations, nonparametric fits are more appropriate, and Gaussian Process Models (GPMs) are effective [12]. We also used `interpolant()` in `R` from the `BACCO` [13] package for GPMs to fit, and results were indistinguishable from the parametric fit shown in Eq. (1).

As mentioned above, Eq. (1) is easily extended to include CR as a fourth predictor. Again we selected only those terms that significantly improved model fit and the resulting model (not shown) accounted for 99.95% of the variance of M_{Pu}, resulting in an RESD of approximately 2% in comparing M_{Pu} to $\hat{M}_{Pu}$. Because Eq. (1) with only 3 predictors (if quadratic and interaction terms are included) explained 99.7% of the variance of M_{Pu}, there was little room for improvement by adding CR to Eq. (1).

There are at least two advantages of a parametric model such as Eq. (1). The first advantage is that variance propagation using the standard “delta method” (linear approximation) is simple and quite accurate. The second advantage is that model discrepancy terms can be easily introduced and characterized by modifying coefficients in Eq. (1). Such model discrepancy terms are discussed in Section 6 where hypothetical real data is collected to detect and estimate model discrepancy.

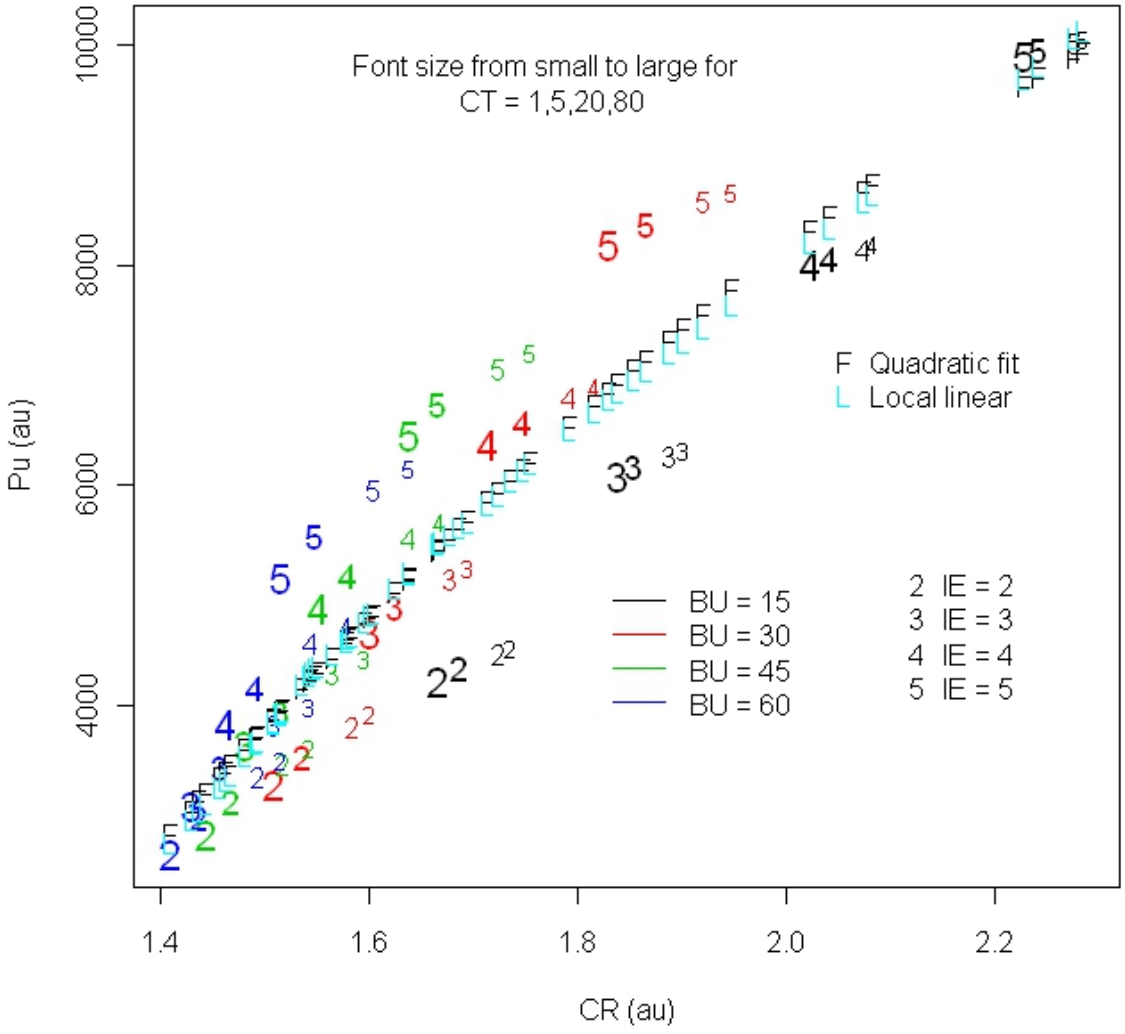

Figure 1. Pu mass in arbitrary units (au) versus the CR for each of the 64 SFAs

The first advantage allows us to confirm our simulation results using the MC (Monte Carlo) approach described in Section 6. The delta method implies that $\sigma_{f_1}^2 \approx \sum_{i=1}^{p} (\frac{\partial f_1}{\partial x_i})^2 \sigma_{x_i}^2$ for a function *f* of variables $x_1, x_2, \ldots, x_p$. Here, f_1 is given in Eq. (1) and x_1 = IE, x_2 = BU, and x_3 = CT. Or, when CR is added, then f_1 is given by the analogous expression as Eq. (1) and x_4 = CR. The partial derivatives are almost trivial to calculate and the delta method is highly accurate because only all third and higher order derivatives are zero and the second order derivatives are relatively small. For example, the delta method RESDs agreed almost exactly with the RESDs from the simulation approach in Section 5 unless the measurement error RESD was 30% or more, in which case the simulation-based RESDs (more reliable in general because the simulation based approach is exact in the limit of running an infinite number of simulations) were approximately 6% higher than the delta method RESDs.

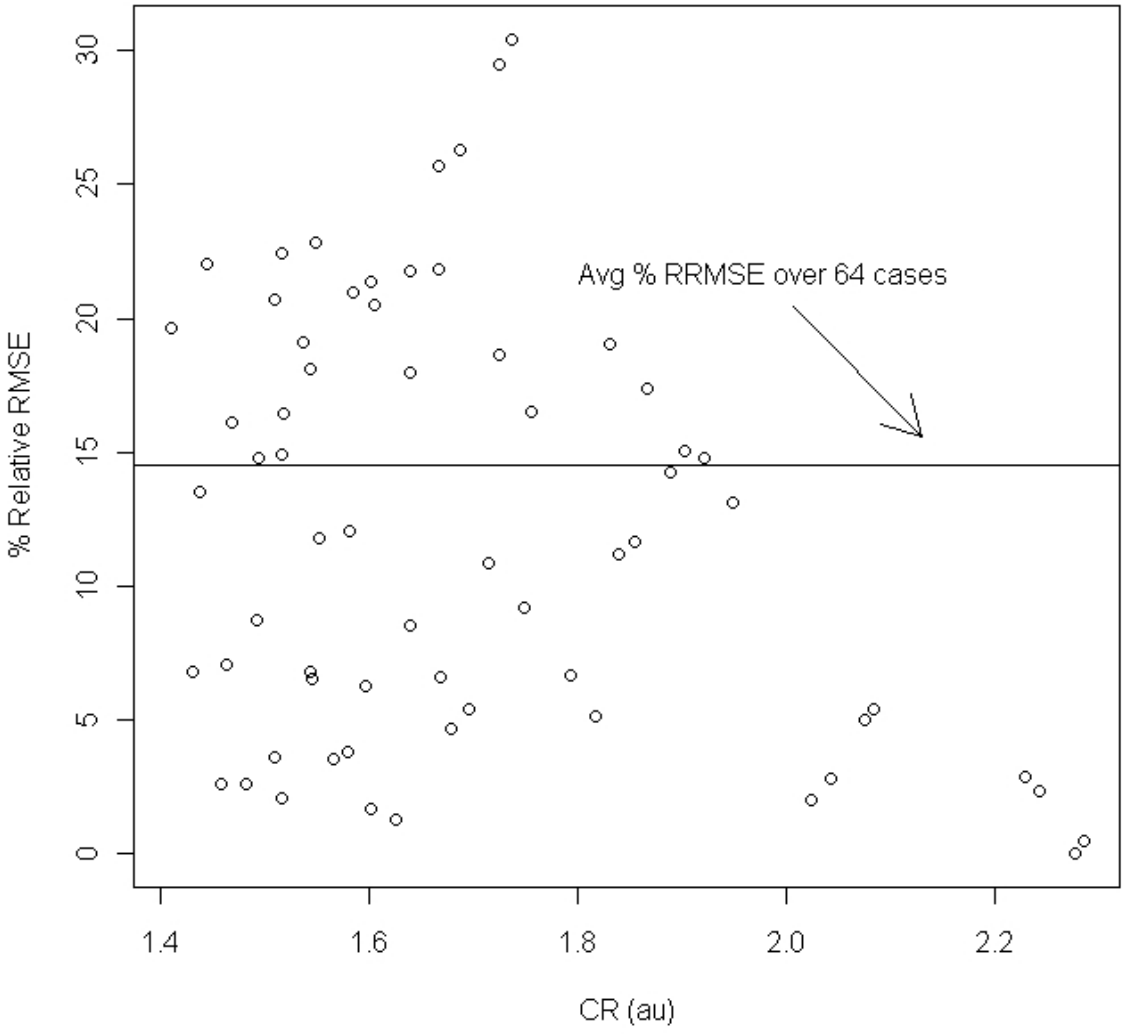

Figure 2 The RMSE in estimated Pu mass in arbitrary units (au) versus the CR for each of the 64 SFAs from Figure 1 The Pu mass was estimated using the parametric fit and zero measurement error.

5. Simulation and MCMC-based Inference Approaches

We include MC simulation and MCMC-based inference approaches to estimate the RESD for each option because both are effective to assess whether using CR to modify the measurement-based estimates of IE, BU, and CT (which are used together with MCNPX to estimate M_{Pu}) improves the RESD. MC simulation is suggested as an alternative to first term Taylor series (the delta method using first derivatives only) in the Guide to expression of Uncertainty in Measurement (GUM) [14]. However, computer model uncertainty brings new challenges not yet addressed in any GUM-like documents.

The MC simulation uses Eq. (2) with f_1 being either the MCNPX output or the fitted output given in Eq. (1). Similarly, a parametric model for CR is also effective and with either f_2 in Eq. (3) being either from the MCNPX output or the fitted model. Eq. (4) is the "prior" probability density for the IE, BU, and CT

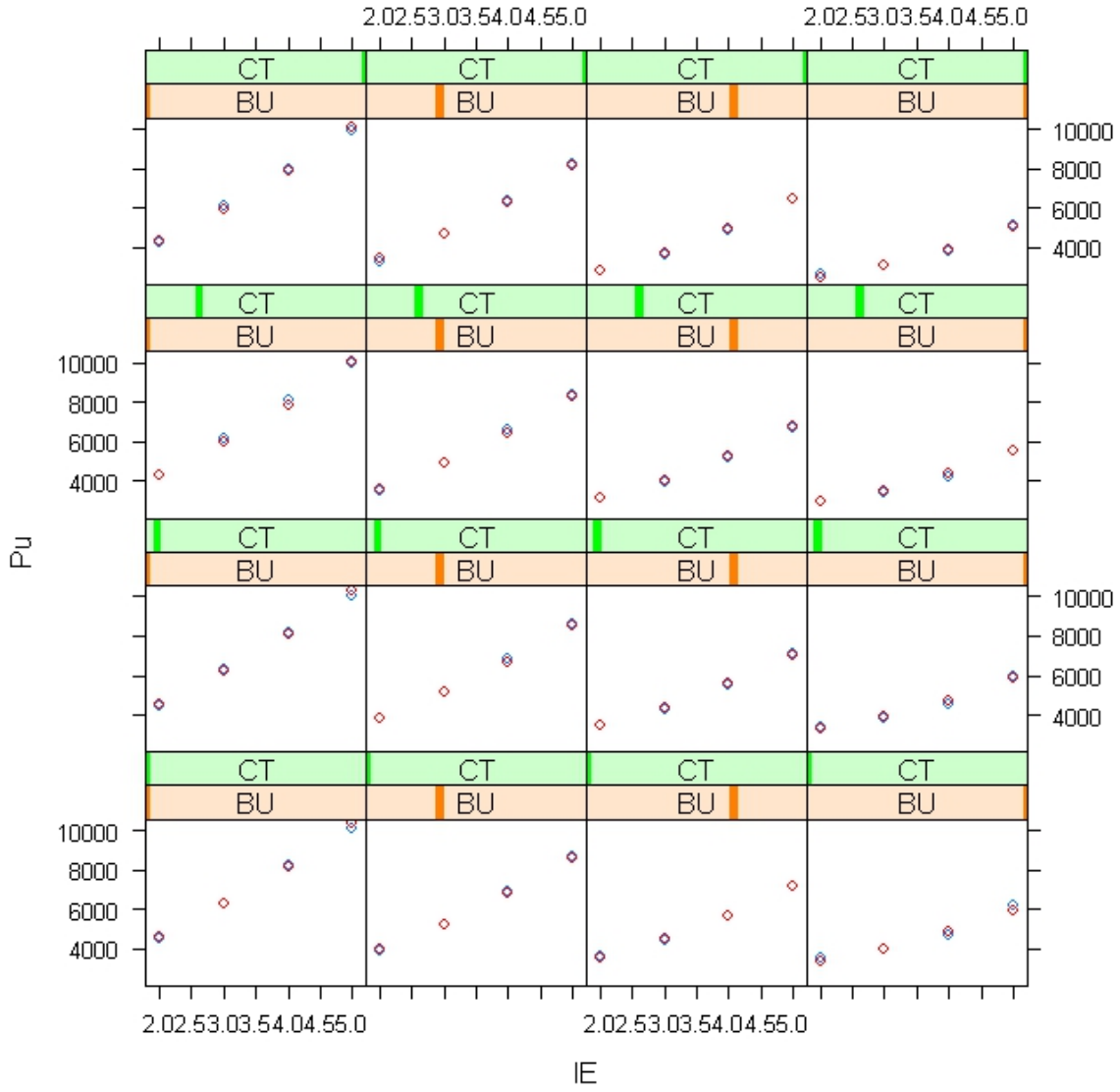

Figure 3. A conditioning plot of the Mass of Pu (au) versus IE for varying values of BU and CT. The values of CT and BU are indicated by the shaded vertical lines. For example, in each row, BU varies from low to high (15, 30, 45, or 60) and CT is 1 in the bottom row, 5 in the next, 20 in the next, and 80 in the top. Two sets of points are plotted. The red points are the MCNPX output. The blue dots (nearly at same locations as the red points) are the fitted values from a simple regression on linear, quadratic, and two interacting linear terms.

values. A uniform prior across the respective ranges can be used. Or, if measurements of any subset of IE, BU, and CT are available, then a normal prior with RESD given by the measurement error RSD can be used. Eq. (4) assumes that prior information about IE, BU, and CT is mutually independent.

$$Pu = f_1(IE,BU,CT) \quad (2)$$

$$CR = f_2(IE,BU,CT) \quad (3)$$

$$p(IE,BU,CT) = p_I(IE)\, p_B(BU)\, p_C(CT) \quad (4)$$

To model the measurement process, assume a true value for IE, BU, and CT. For convenience and as an accepted cross-validation (CV) strategy, we chose i = 1, 2, …, 64 in the 64-design space and then report average RESDs over the 64 possible true values. For a given true IE, BU, and CT, add measurement error as described in Section 3. For each realization of the measurement error process, compute CR from Eq. (3) and the realized values of IE, BU, and CT. Because the RESDS are fairly small (ranging from 1% to 20%), the realized values will be near the true value.

To combine the estimated Pu from each realized value of IE, BU, and CT, [2] suggests a weighted average, with weights $e^{-\frac{\{CR-f_2(\mathrm{IE,BU,CT})\}^2}{2\sigma_{CR}^2}}$ where σ_{CR}^2 is the absolute (not relative) SD of the CR measurement. Here is where code emulation enters. Recall that only the 64 combinations of IE, BU, and CT values are available. For other values, interpolation is needed. Eq. (1) in Section 3 makes the interpolation simple. GPMs are also very suited for interpolation. Both options can include uncertainty in the interpolation. However, in this context including interpolation uncertainty requires an extension that is under development. Therefore, we only consider interpolation uncertainty using uncertainty in the parameter fits from Eq. (1). The end result for the MC simulation based estimate of M_{Pu} can then be written as

$$\hat{M}_{\text{Pu by simulation}} = \frac{\sum_{i=1}^{n} e^{-\frac{\{CR-\hat{f}_2(\mathrm{IE_i,BU_i,CT_i})\}^2}{2\sigma_{CR}^2}} \hat{f}_1(IE_i, BU_{\mathrm{i}}, CT_{\mathrm{i}})}{\sum_{i=1}^{n} e^{-\frac{\{CR-\hat{f}_2(\mathrm{IE_i,BU_i,CT_i})\}^2}{2\sigma_{CR}^2}}} \qquad (5)$$

where *n* is the number of simulations, say 1000 or more. The *n* simulations model possible measurement results for IE, BU, and CT, so IE, BU, and CT have the subscript *i* inside the summation in Eq. (5). The simulation approach intends to mimic a situation in which there is only one measurement of each of IE, BU, CT, and CR. Therefore, summation over the *n* simulations requires explanation. First, the Gaussian-style weights are reasonable, but ad hoc at this stage. They can be formally defended by adopting a likelihood view as described next.

For MCMC-based inference**,** Eq. (4) is the "prior" probability density for the IE, BU, and CT values. A uniform prior across the respective ranges can be used. Or, if measurements of any subset of IE, BU, and CT are available, then a normal prior with RESD given by the measurement error RSD can be used to generate the *i* = 1, 2, …, *n* values $IE_i, BU_{\mathrm{i}}, CT_{\mathrm{i}}$ used in Eq. (5). Because of the term $e^{-\frac{\{CR-f_2(\mathrm{IE,BU,CT})\}^2}{2\sigma_{CR}^2}}$, which results from comparing Eq. (3) to the measured CR , Eq. (5) is then related to the "posterior" probability density for IE, BU, and CT, Π(IE,BU,CT), which via Eq. (3) gives the posterior distribution for M_{Pu} . In that way, the measured CR is allowed to improve/update the prior probability density for IE, BU, and CT in going from Eq. (4) to Eq. (5). Because we assume the prior information and measurement errors in IE, BU, and CT are independent,

$$\Pi(\mathrm{IE,BU,CT}) = \pi_{\mathrm{I}}(\mathrm{IE})\, \pi_{\mathrm{B}}(\mathrm{BU})\, \pi_{\mathrm{C}}(\mathrm{CT}) \qquad (6)$$

More formally, and leading to MCMC, we have two possible priors (prior to observing CR) as follows.
Prior 1:
Sample IE, BU, CT independently from uniform distributions spanning the ranges 1-5 for IE, 15 to 60 for BU, and 1-80 for CT, or

Prior 2:
Sample IE, BU, CT independently from normal distributions centered on the true IE, BU, CT values with standard deviations of approximately 1% to 20% relative to the true values.

Then, the prior distributions 1 or 2 for IE, BU, and CT can be updated to form posterior distributions given a measurement of CR by using Eq. (3) and assuming the CR is Gaussian distributed to compute the likelihood. To do so, we use MCMC to numerically obtain samples from Π(IE,BU,CT), the posterior distribution of IE,BU,CT. Because the term $e^{-\frac{\{CR-f_2(\mathrm{IE,BU,CT})\}^2}{2\sigma_{CR}^2}}$ is the only part of the Gaussian

likelihood that matters in this context, the intuitive weighting scheme described in the MC simulation paragraph is very similar to the MCMC scheme. The difference is that in MCMC, trial moves are made to change IE, BU, and CT values and these trial moves are accepted with a probability that depends on the prior *x* likelihood. In the MC simulation, all simulated values of IE, BU, and CT are used, but those that do not fit CR well are downweighted in $\hat{M}_{\text{Pu by simulation}}$ given in Eq. (5)

Although we do not think there are important differences in the two methods in this context, we implemented both and compared the estimated RESDs for $\hat{M}_{\text{Pu by simuilation}}$ to those for $\hat{M}_{\text{Pu by MCMC}}$. In most examples considered, the estimated RESDs are indistinguishable. However, some results are given in Section 6 where for large error variances, there are reproducible differences in the two methods. To implement MCMC in this context we used `metrop` from the `BACCO` package in `R` [15].

As a final point to make, Eq. (5) uses $\hat{f}_1$ and $\hat{f}_2$ to denote estimates of the corresponding true f_1 and f_2. Because MCNPX is run to compute f_1 and f_2 only at the IE, BU, and CT values, some type of interpolation is required. This is another advantage of parametric fitting such as Eq. (1). We have also used kernel smoothing, local linear smoothing, and `interpolant()` in `R` which implements GPMs. Results are reported in section 6 for parametric fitting. Results are very similar for these nonparametric fits, although in general there could be different RESDs for the various fitting options.

6. Example RESD results

Table 1 gives RESD results for a few cases. Cases 2-5 have two entries. For cells having two entries, the first entry includes emulation/interpolation error and the second entry omits it. In this context, and using parametric fitting code emulation error appears to be small. Recall that results for the GPM fitting were very close to those from parametric fitting so were not included.

The Table 1 cell entries first ignore emulation error by assuming the fitted M_{Pu} from Eq. (1) are the true values, and then include emulation error by using the MCNPX output values for M_{Pu}, in which case the emulation RESD is approximately 2% (see Section 3 just below Eq. (1)) in comparing M_{Pu} to $\hat{M}_{Pu}$. Related to emulation error is the tendency for any method to overfit training data. Therefore, Eq. (1) was applied with each of the 64 sets of parameter values being omitted in turn, which is known as leave-1-out cross validation (CV) [11]. However, RESD values with and without CV are similar. All shown RESDs include CV.

Case 1 is the 14% average RESD over the 64 cases described in Section 4, but with measurement error effects added. As described in Section 3, measurement errors are random with relative standard deviation σ_{RSD}.

Case 2 uses only IE, BU, and CT via Eq. (2) and $\hat{M}_{\text{Pu by simulation}}$.

Case 3 uses IE, BU, CT, and CR via the parametric fit. Recall that we also considered simply adding CR as a fourth predictor. This seems to be ineffective, with large RESDs unless the σ_{RSD} for CR is kept to a low value such as 0.01 as in case 3b. That is, Table 1 entries for case 3b keep for CR at 0.01 across all columns but vary σ_{RSD} for IE, BU, and CT across the columns. If the σ_{RSD} for CR varies across the columns (Case 3b) as it does for all cases except 3a, then RESD values are very large, as shown.

Case 4 uses CR to weight the proposed/measured IE, BU, CT values.

Case 5 is the same as case 4 but uses the prior/posterior formulation and MCMC. The RESDs for each case are given in Table 1 for measurement RESDs of 1%, 2%, 5%, 10%, and 20%.

Because the role for CR in case 5 is to update the prior in Eq. (4) to the posterior in Eq. (5), it is useful to assess the standard deviation of IE, BU, and CT in the prior and in the posterior. As a rough summary, if σ_{RSD} for CR is 1%, then the posterior standard deviation of IE is reduced substantially, is only slightly reduced for BU and CT. Recall from the sensitivity discussion in Section 4 that CT does not have much impact on the overall RESD and neither does BU if DR is used.

Table 1. M_{Pu} estimation % RESDs for cases 1-5 for measurement RESDs. For cells having two entries, the first entry includes emulation/interpolation error and the second entry omits it. For case 3, the % RESDs are much higher (case 3a) if the σ_{RSD} for CR is the same as the RESD for IE, BU, and CT. Entries in the table for case 3b are for σ_{RSD} = 1% for CR and 1%, 2%, 5%, 10%, or 20% for each of IE, BU, and CT.

Case	Measurement σ_{RSD}				
	1%	2%	5%	10%	20%
1: CR only	15	17	25	41	71
2: IE BU, CT	3,1	3,2	6,5	11,10	22,21
3a*:IE,BU,CT,CR*	2,2	4,4	10,10	22,22	46,46
3b*:IE,BU,CT,CR*	2,1	3,2	5,5	10,10	21,21
4: CR to weight	3,1	3,2	4,4	10,10	19,19
5: CR in MCMC	2,2	3,2	4,4	7,7	12,12

7. MCNPX "calibration"

All previous sections have ignored MCNPX model error. Work is ongoing to improve the MCNPX modelling but all models are intentional simplifications of complex phenomenon and so are "wrong." Choosing the appropriate model fidelity by including features such as shuffling assemblies during their stages in the reactor, loading poisons and stratifying enrichments is being considered. A key question is what model fidelity is required to effectively assess NDA options and associated analyses. Until those efforts are further along, we will simply consider using actual measurement results of Pu to adjust the MCNPX output.

As a first step to assessing sample sizes required to "adjust/calibrate" MCNPX output in Eqs. (2) and (3), we introduced MCNPX model error by modifying in turn each of the 9 coefficients in Eq. (1), resulting in 9 types of model discrepancy. The modification amounts were chosen using an optimizer (`optim`) in `R` so that the average relative standard deviation of the discrepancy between the true response and the MCNPX modelled response was 5%. We then randomly selected 2, 4, 8, 16, 32, 48, or 64 of the 64 design points to be measured. The modelled response was then modified by using the measured rather than the predicted value. Future work will combine both the measured and predicted values to improve the initial prediction. At this stage, despite errors in the measurements, the measured results are thought to be closer to the true values than the modelled results.

Somewhat surprisingly, the effect of the 5% relative error in MCNPX model predictions was substantially present (in terms of the average bias across the 64 design points) for each of the 9 discrepancies until there were at least 48 calibration points. Specifically, the average % relative bias across the 64 design points for each of the 9 discrepancies range from 3 to 5, 2 to 5, 2 to 5, 2 to 4, 1 to 3 and 1 to 1 for 2,4,8,16,32, and 48 samples, respectively. The average % relative bias without any discrepancy term was 0 without emulation/interpolation error and 0.1 with interpolation error.
We caution that this pessimistic result is not a general result, but it does apply to the example PNAR-type response used here for the 9 types of model discrepancies considered.

The simpler case of pure "white noise" model error is easily accommodated by adding the variance of the model error to the variances (which underly the Table 1 entries for example) reported here. Independent white noise model error could arise as "assembly-specific" modelling errors, unlike the 9 model error types just described.

8. Summary

We considered how to use one modelled detector response (DR) to improve estimation of Pu mass (M_{Pu}). Error sources considered included model output emulation/interpolation errors and measurement error models. Current measurement error models assumed that errors in IE, BU, and CT are independent. Initial steps to include structured model error were briefly described in Section 7. Additional work on assessing code emulation error in this context using GPMs is underway.

9. Acknowledgements

We acknowledge the Next Generations Safeguards Initiative within the US Department of Energy.

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LA-UR 11-02596

Fissile Isotope Discrimination in Spent Fuel Assemblies by Analysis of the Correlated Neutron Signal

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Abstract:

The Next Generation Safeguards Initiative (NGSI) of the U. S. Department of Energy currently supports a substantial research effort focused on quantifying plutonium (Pu) mass in spent fuel using non-destructive assay (NDA) techniques. The Differential Die-Away Self-Interrogation (DDSI) technique, one of the several being investigated, has been shown to quantify the total fissile content in spent fuel. Unlike active-interrogation NDA methods, DDSI does not require an external neutron source, but rather, uses spontaneous fission neutrons within the spent fuel for self-interrogation. The essence of the technique lies in the time separation between the detection of spontaneous fission neutrons, captured in a signal-triggered early gate, and the capture of the dominant fraction of induced fission neutrons during a later gate, thus enabling the independent measurement of fissile and fertile mass in the sample. The time domain analysis of the doubles distribution also reveals a difference in the temporal fission response of ^{235}U compared to ^{239}Pu, due to their differing higher order induced fission moments. We have simulated the behaviour of a prototypical DDSI instrument using MCNPX to model the detector response for a library of PWR fuel assemblies. Also, by exploiting the recent advancements in the Particle Track (PTRAC) capability in MCNPX, it is possible to generate a capture time distribution with isotopic designation to partition the temporal signal from individual isotopes. Although PTRAC distributions from individual isotopes are not experimentally verifiable, the information they provide is used to optimize experimental acquisition gates for time-correlated counts to capture temporal differences from each isotope. Here, we explore the use of a previously established discrimination approach, the prompt-to-delayed response ratio, applied our generated spent fuel library. We then investigate the feasibility of using this novel time correlation analysis by comparison to the prompt-to-delayed approach.

Keywords: spent fuel; plutonium; nuclear safeguards; non-destructive assay

1. Introduction

Within the nuclear safeguards framework, there is great demand for the development of NDA instruments capable of measuring the fissile materials in Pu/U mixed fuels, such as spent fuel. The high gamma fields and large neutron backgrounds in spent fuel preclude the measurement of uranium and plutonium by conventional passive or active methods. For high burnup LWR fuel, there are three major fissile isotopes (^{235}U, ^{239}Pu, ^{241}Pu) which all contribute toward the detected signal. These isotopes vary with burnup, initial enrichment and cooling time. In order to quantify plutonium mass in spent fuel, we must be able to quantify the contribution of the individual fissile isotopes.

A few techniques capable of measuring ^{239}Pu and ^{235}U in mixed fuel have been investigated. One common approach involves the measurement of both the prompt and delayed fission neutron response. The prompt-to-delayed response ratio is then used to separate the ^{239}Pu and ^{235}U contributions. This approach has been demonstrated for fresh BWR rods [1] and fresh FBR, ATR, and LWR fuel rods, as well as irradiated LWR fuel pellets [2]. In this paper, we investigate the feasibility of using the prompt-to-delayed response ratio for the determination of ^{239}Pu and ^{235}U content in

simulated spent PWR assemblies. We use previously generated prompt and delayed neutron response data from the Differential Die-Away Self-Interrogation technique (DDSI) [3] and the Delayed Neutron [4] instrument, respectively, both of which have been studied as a part of the NGSI effort [5]. Other neutron techniques, such as the lead slowing down spectrometer (LSDS) and the Neutron Resonance Transmission Analysis (NRTA) may also provide this information. LSDS distinguishes the fissile isotopes based on cross-section differences at certain energies using time-energy correlation. This approach has been investigated for fresh and irradiated fuel rods [6], as well as for PWR spent fuel assemblies [7]. NRTA exploits the individual resonance absorption lines of the various fissile isotopes present in the fuel [8].

Here, we investigate the feasibility of using the correlated neutron signal obtained from the Differential Die-Away Self-Interrogation (DDSI) for the purpose of fissile isotope discrimination in PWR spent fuel assemblies. The DDSI instrument is shown in Figure 1. The technique uses the spontaneous fission neutrons from ^{244}Cm within the assembly as the "pulsed" neutron source. The time correlated neutrons from the spontaneous fission and the subsequent induced fissions are analyzed as a function of time after the trigger event to determine the spontaneous fission rate and the induced fission rate in the sample. Fissile mass is determined from the count rate acquired during the late gate. The dominant fraction of induced fission occurs later in time with neutrons that have been moderated and reflected toward the assembly. The doubles in the late gate, D(100-196μs), divided by the singles, S, (ratio denoted as *D(100-196)/S*) is used to determine the fissile content.

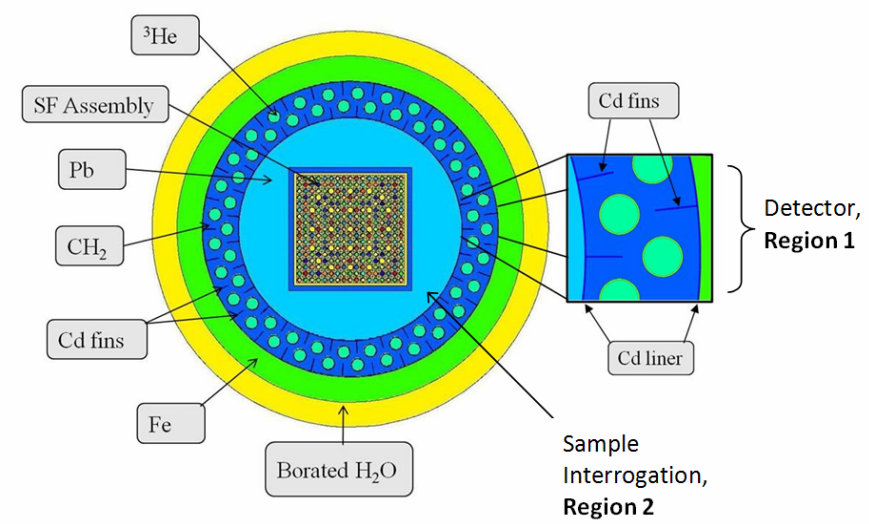

Figure 1: A horizontal cross-section of the DDSI detector configuration for MCNPX simulations is shown.

The DDSI technique has been used to quantify the fissile content in a range of possible PWR spent fuel assemblies. A comprehensive assembly library was developed [9] to provide a standard measurement sample set for the several NDA techniques being investigated in this research effort. The entire library contains 64 PWR spent fuel assemblies with burnup values of 15, 30, 45 and 60 GWd/tU; initial enrichment values are 2, 3, 4, 5 wt% ^{235}U, and cooling times are 1, 5, 20, and 80 years. For the prompt-to-delayed analysis presented in section 2, only the 5-yr cooled assemblies are considered (sixteen cases). For the time correlation analysis presented in section 3, we consider only eleven assemblies, a subset of the sixteen cases mentioned above, where the improbable cases of 2% and 3% both at 45 GWd/tU and 60 GWd/tU, and 4% at 60 GWd/tU have been removed.

2. Fissile isotope discrimination using the prompt-to-delayed response ratio

Active interrogation NDA techniques, which measure the prompt induced fission response, as well as the delayed fission response, have been used to verify both the uranium and plutonium content of spent LWR fuel. The prompt/delayed-neutron ratio is used to "separate" the contribution from ^{235}U and ^{239}Pu. The reason why the prompt/delayed-neutron ratio depends on the U/Pu ratio is due to the fact that ^{235}U and ^{239}Pu have very differing delayed-neutron yields, i.e. the delayed neutron yield of ^{235}U is

2.56 times larger than that of ^{239}Pu. The prompt-delayed method has not been used in the presence of ^{241}Pu, which is present in high burn-up fuel. This concern will be addressed later.

In general, pulsed-neutron or shuffled isotopic sources are used to interrogate the fuel, and the prompt-neutron measurement time is chosen to capture the fissile content contribution, and the delayed-neutron measurement, which is taken while the source is "off". The total fissile content response can be captured using techniques, such as DDSI, Differential Die-Away Analysis (DDA) [10], ^{252}Cf Interrogation Prompt Neutron (CIPN) [11], and other techniques which have been investigated under this research effort.

In practice, only one detector configuration should be used to measure both the prompt and the delayed signal; however, we will use the prompt signal from DDSI and the delayed signal from a separate delayed neutron (DN) instrument by Blanc et. al. [4], since the data is already available; even though the DDSI and DN are not hardware-compatible. Here we apply the prompt-delayed technique for our simulated assemblies in order to: (1) ensure that the physics of fissile isotope discrimination have been captured in our simulations, and (2) to use the prompt-delayed discrimination as a basis for comparison for the time-correlated discrimination, developed in the subsequent section.

2.1. The two-component linear system

The two-component linear system, as defined by Equation 1, is used as a starting point for ^{235}U and ^{239}Pu discrimination, meaning the presence of ^{241}Pu is ignored. The prompt and delayed neutron response, P and D respectively, are shown in Equation 1, as suggested by Matsuda et. al. [2],

$$P = a_1 N_5 + a_2 N_9 \qquad \text{Eq. 1a}$$

$$D = b_1 N_5 + b_2 N_9 \qquad \text{Eq. 1b}$$

The N_5 and N_9 values are the masses of ^{235}U and ^{239}Pu, respectively, in the interrogated section of the fuel assembly, while the a_1 and a_2 coefficients weigh the prompt neutron response per unit mass of ^{235}U and ^{239}Pu respectively. Likewise the b coefficients weigh the delayed neutron response for each isotope. Note that the P value in Eq. 1a will be represented by the DDSI ratio, i.e. the D(100-196μs)/S ratio, normalized to the assembly of least fissile content. Likewise, the D values in Eq. 1b are normalized to the least fissile assembly; therefore the D/P ratio is dimensionless. Note that the P and D values are obtained from separate instruments with different interrogating source strengths, but the normalized values are independent of the source strengths. The normalized D/P ratio shown in Figures 2 and 3 merely captures the relative change of the D/P ratio among the sixteen assemblies and does not show the actual delayed-to-prompt ratio for a given assembly. We use Eq. 1, expressed in the matrix algebra form of Eq. 2a, to define a discrimination ratio, DR, a dimensionless parameter used to quantify the discrimination ability of the two-component system; where $X=\begin{pmatrix} P \\ D \end{pmatrix}$ and $N=\begin{pmatrix} N_5 \\ N_9 \end{pmatrix}$. This system will only have a solution if the determinant of the coefficient matrix is not equal to zero, i.e. if the coefficient matrix has an inverse, as shown in Eq. 2b.

$$X = \begin{bmatrix} a_1 & a_2 \\ b_1 & b_2 \end{bmatrix} \cdot N \qquad \text{Eq. 2a}$$

$$N = \begin{bmatrix} a_1 & a_2 \\ b_2 & b_2 \end{bmatrix}^{-1} \cdot X = \frac{1}{|(a)|_1 b_2 - a_2 b_1)} \begin{bmatrix} b_2 & -a_2 \\ -b_1 & a_1 \end{bmatrix} \cdot \begin{pmatrix} P \\ D \end{pmatrix} \qquad \text{Eq. 2b}$$

Therefore if $a_1b_2=a_2b_1$, there is no solution and hence no discrimination; so if DR is defined as (a_1b_2/ a_2b_1), there is no discrimination if DR is equal to unity. DR should be much smaller than unity, with the smaller DR value translating to better fissile isotope discrimination. For DDSI [12], the average a_1=0.46; a_2=1 (w.r.t. to ^{239}Pu) and for DN [13], the average b_1=1.54 and b_2=1, so a prompt-delayed system using DDSI and DN has a DR=0.30. The DR values for other prompt-delayed integrated systems are shown in Table 1.

Integrated System	Discrimination Ratio (DR)
DDSI+DN	0.30
CIPN+DN	0.34
DDA+DN	0.35

Table 1: DR values for ^{239}Pu and ^{235}U separation for prompt-delayed integrated NDA systems.

Figure 2 shows the D/P ratio for the sixteen assembly cases (listed in Table 2) versus the ^{239}Pu/(^{239}Pu+^{235}U) mass ratio using the DDSI and DN data.

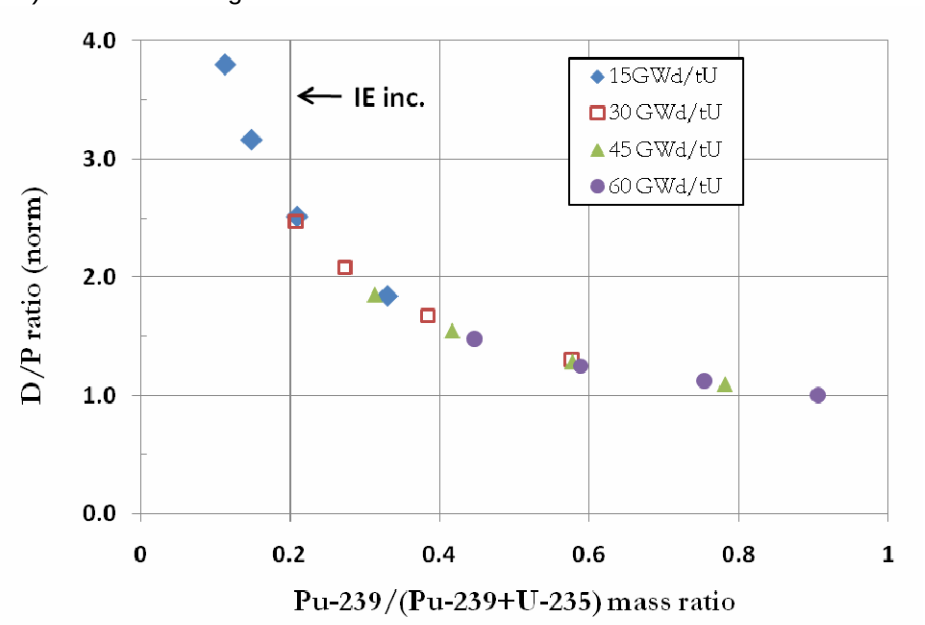

Figure 2: Correlation between the D/P ratios and the ^{239}Pu/(^{239}Pu+^{235}U) ratios for 16 assemblies

CT [yr]	BU [GWd/tU]	IE [wt% ^{235}U]	^{239}Pu/(^{239}Pu+^{235}U)
5	15	2	0.33
5	15	3	0.21
5	15	4	0.15
5	15	5	0.11
5	30	2	0.58
5	30	3	0.38
5	30	4	0.27
5	30	5	0.21
5	45	2	0.78
5	45	3	0.58
5	45	4	0.42
5	45	5	0.31
5	60	2	0.91
5	60	3	0.75
5	60	4	0.59
5	60	5	0.45

Table 2: Sixteen assembly cases used in analysis and corresponding ^{239}Pu mass ratio [row in red corresponds to maximum 239Pu mass fraction, while blue indicates least]

Note that there is a clear correlation between the D/P ratio and the $^{239}Pu/(^{239}Pu+^{235}U)$ ratio regardless of the initial enrichment and burn-up of the assemblies. In other words, the combination of the simulated prompt neutron signal from DDSI, which captures the fissile content, and the simulated delayed neutron signal from the DN instrument, provides the necessary information for fissile isotope discrimination; the physics necessary for discrimination has been captured in our simulations. If the prompt and delayed signals are accurately scaled based on the interrogating neutron source strengths and the integrated system is calibrated using a standard sample, the correlation seen in Figure 2 can be used to determine the ^{239}Pu and ^{235}U mass ratio quantitatively. The correlation, however, does not appear to be linear, as one would expect given the defining relation stated in Eq. 1. Figure 3 shows this deviation from linearity, with the greatest deviation occurring at low burn-up highly enriched fuel, where the ^{235}U is greatest component of the fissile content. The linear response is calculated using Eq.1 with the weighting coefficients tallied from the MCNPX simulation. The "detected" response is also obtained from the simulated prompt and delayed counts arriving at the detector. This discrepancy shows that the linear two-system model, although it shows the negative correlation between the D/P ratio and the ^{239}Pu mass fraction, does not adequately characterize the system, with the ^{241}Pu contribution being ignored.

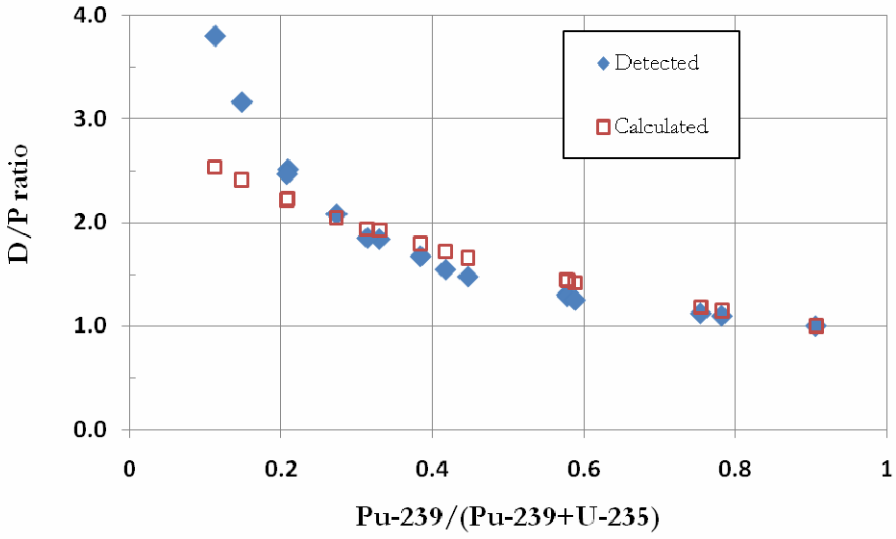

Figure 3: Detected and calculated correlation between the D/P ratios and the $^{239}Pu/(^{239}Pu+^{235}U)$ ratios for 16 assemblies

2.2. ^{241}Pu Correction

Among the sixteen assemblies simulated, the ^{241}Pu content can be as high as 20% of the total fissile content, so Eq. 1 does not adequately describe the prompt and delayed fission response. It is not possible to quantify isotope discrimination ability in a three-component system, so the analysis presented above cannot be simply extended by adding a third term, for ^{241}Pu, to Eq. 1a and 1b. Also of note is the fact that ^{241}Pu has a delayed neutron fraction similar to that of ^{235}U (0.0154 vs. 0.0158 respectively), so as the ^{241}Pu increases, the discrimination between ^{235}U and Pu will diminish. One method of correcting for the presence of ^{241}Pu is to use an iterative scheme, as proposed by Menlove et. al.[14], where a two-component system with ^{239}Pu and ^{235}U is initially assumed (no ^{241}Pu present). The D/P ratio is used to obtain a first guess of the ^{239}Pu to ^{235}U ratio. Based on this first guess, isotopic correlation would then be used to estimate ^{241}Pu content. This estimate of ^{241}Pu mass is then used to correct the ^{239}Pu and ^{235}U ratio, which will then be used to update the ^{241}Pu mass, until convergence occurs for all three components. It is clear that either isotopic correlation or information from another NDA measurement is needed to de-convolute the contribution of all three major fissile isotopes i.e. three observables are needed to solve for three unknowns. This will be the subject of future work.

3. Fissile isotope discrimination using the correlated neutron signal

The detected induced fission response as a function of time depends on the relative concentrations of ^{239}Pu and ^{235}U. Table 3 shows the induced fission moments of these isotopes for the sake of comparison. Note that ^{239}Pu has increasingly larger second and third induced fission moments than ^{235}U, meaning the time correlations of the induced fission neutrons can give the relative concentrations of the two isotopes.

Isotope	ν	$\nu(\nu-1)$	$\nu(\nu-1)(\nu-2)$
^{235}U	2.406	4.626	6.862
^{239}Pu	2.879	6.733	12.630
Ratio ^{239}Pu/^{235}U	1.197	1.455	1.84

Table 3: Prompt fission neutron multiplicity moments for ^{239}Pu and ^{235}U

The two isotopes also differ in the energy-dependence of their fission cross-section in the epi-cadmium region, with ^{239}Pu fission occurring preferentially over ^{235}U fission at the 0.3 eV resonance energy. For the single fuel pin assay, this cross-section difference at a specific energy results in a time signature in the response, as the neutron must first slow down to that energy (time-energy correlation). This signature is useful for the assay of a single fuel pin, which is compact with low multiplication, but for the assay of a large 16x16 assembly submerged in water, the high multiplication smears this distinct time feature, and the increased size of the sample diminishes the energy resolution.

We can sample the induced fission rate as a function of time after the first detected neutron using list-mode data collection. A measure of the die-away time of these correlated distributions should depend on the relative concentrations of ^{239}Pu and ^{235}U. The doubles and triples distribution for ^{239}Pu should peak earlier and have a shorter die-away time than that of ^{235}U. This would be observable experimentally if two separate samples with only either of the isotopes present (no mixture-pure sample), but this cannot be readily observed in a mixed sample since we do not know the isotopic origin of the detected fission neutrons. A recent modification to the particle track (PTRAC) capability in MCNPX [15] breaks down the isotopic contributions such that this effect can be observed, and furthermore used to optimize gates to determine the relative concentrations of the two isotopes. Figure 4 shows the results of the PTRAC analysis of a simulated 45GWd/tU, 4 wt% ^{235}U, and 5-yr cooled assembly.

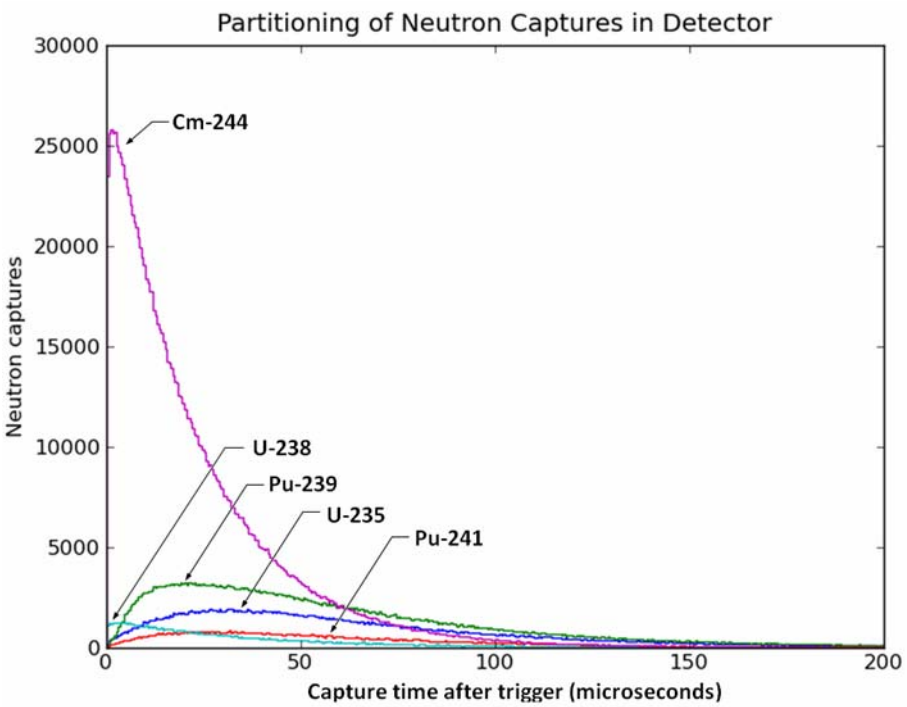

Figure 4: The enhanced PTRAC Capture file capability labels the isotopic origin of each detected neutron. Neutrons are captured in ^{3}He detectors in the DDSI geometry surrounding a 45GWd/tU, 4% IE, and 5-year cooled PWR assembly.

The total detected induced fission neutron response is determined by the time response of the three major fissile isotopes, ^{239}Pu, ^{235}U, and ^{241}Pu, The shape of the total induced fission time response will depend on the relative concentrations of these isotopes. The PTRAC capability can also sum the total induced fission time response, as shown in Figure 5. Here, the normalized induced fission distributions are shown for assemblies with the smallest and largest ^{239}Pu fraction across the 11 assemblies listed in Table 4. The curves are normalized to only show shape differences in the distributions. The smallest and largest ^{239}Pu fraction cases define an envelope of variation of the induced fission response as seen in Figure 5. These two distributions contain valuable information for doubles gate optimization in DDSI to capture this shape difference and hence the relative concentration of the two isotopes.

CT [yr]	BU [GWd/tU]	IE [wt% ^{235}U]	^{239}Pu/(^{239}Pu+^{235}U)
5	15	2	0.33
5	15	3	0.21
5	15	4	0.15
5	15	5	0.11
5	30	2	0.58
5	30	3	0.38
5	30	4	0.27
5	30	5	0.21
5	45	4	0.42
5	45	5	0.31
5	60	5	0.45

Table 4: Sixteen assembly cases used in analysis and corresponding ^{239}Pu mass ratio [row in red corresponds to maximum mass ratio, while blue indicates the least]

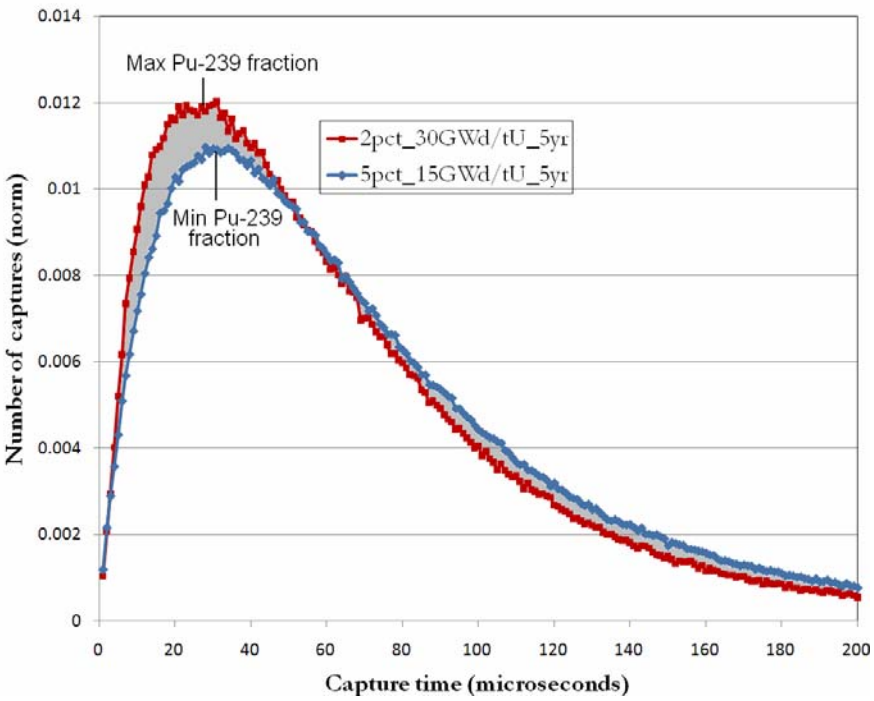

Figure 5: Normalized Induced fission distribution envelope defined by the maximum and minimum ^{239}Pu fraction assemblies obtained from the PTRAC capture file in MCNPX simulation (for 11 assemblies considered)

As seen from Figure 5, the ^{239}Pu fraction affects the rise time and the die-away time of the neutron capture distributions, obtained using the PTRAC capture file in MCNPX. The shape of the capture distributions in Figure 5 can be characterized by the R-parameter, defined in Equation 3 and shown in Figure 6. Here, R is defined as the ratio sum of the captures in the "tail" to the total number of captures over all time, in essence the ratio of the tail integral to the total integral. The R-parameter was chosen to characterize the shape of the capture distribution borrowing from a common pulse shape discrimination method in detector physics.

$$R = \frac{Tail}{Total} \qquad \text{Eq. 3}$$

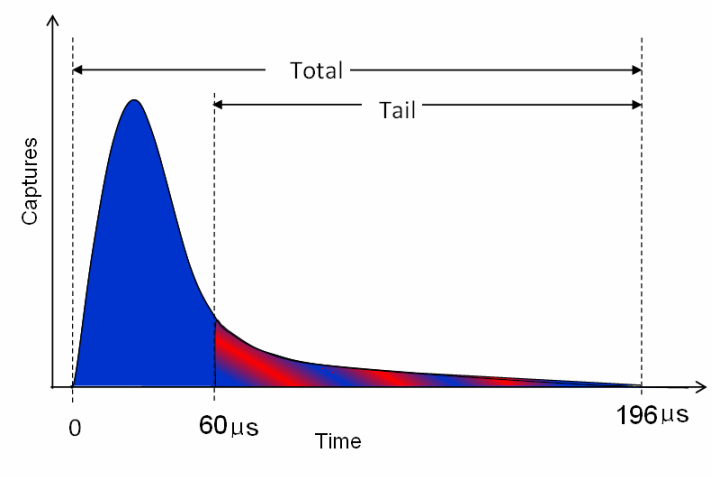

Figure 6: R-parameter used to characterize shape of the capture distribution as a function of time with a 4 μs gate width

A similar characterization can be performed using the factorial moment distribution, proportional to the doubles, from the F8 Capture tally capability of MCNPX. Figure 7 shows the dependence of the factorial moment (and hence the doubles) as a function of pre-delay after the trigger event for the limiting cases shown in Table 4, A similar shape difference, as that seen in the capture distribution is observed.

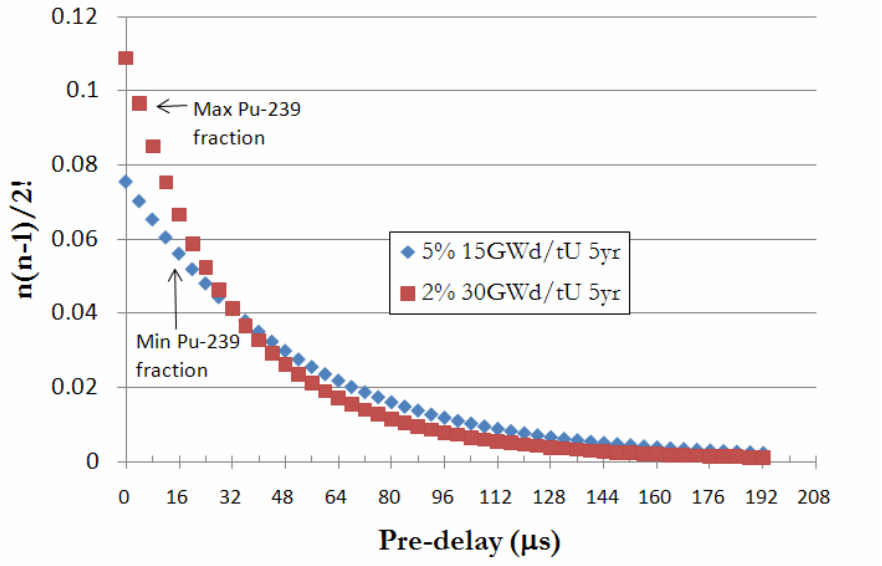

Figure 7: Normalized factorial moment distribution for limiting cases with max and min ^{239}Pu fraction with a gate width of 4μs

The R-parameter can be re-defined by Eq. 4 to be used for Figure 7.

$$R = \frac{D(60-196)}{D(0-196)} \qquad \text{Eq. 4}$$

By inspection of Figure 5, the tail start time was chosen as 60 μs and the overall time extended to 196 μs. In conventional doubles counting, the tail gate can be optimized to detect this change in shape of the doubles distribution, and hence the fissile Pu/U ratio.

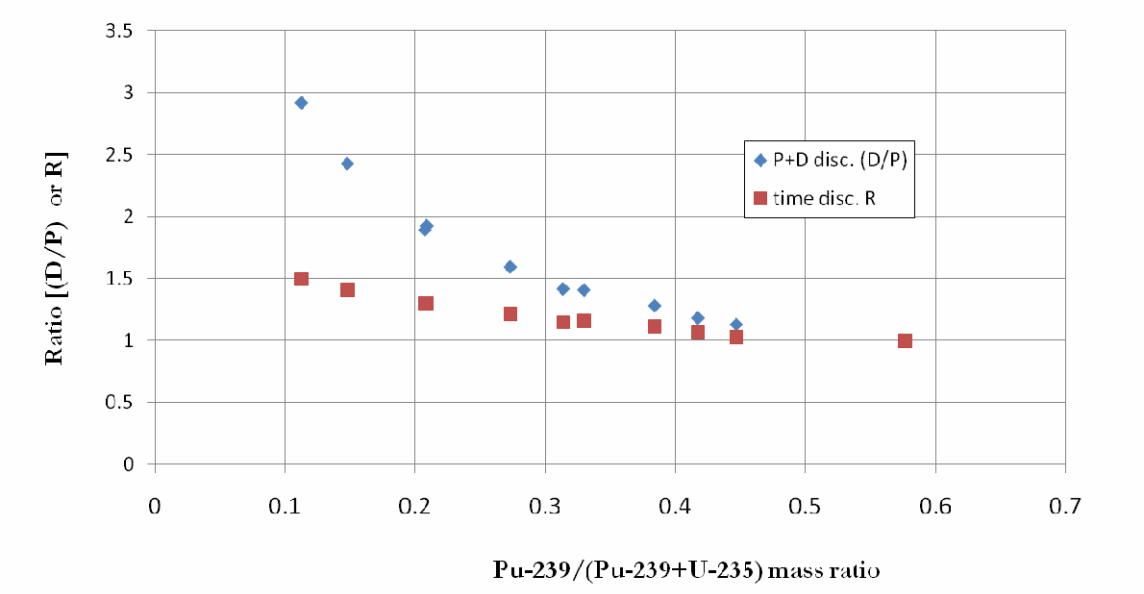

Figure 8: R shape parameter and the D/P ratio for 11 assembly cases as a function of fissile Pu/U mass ratio

Figure 8 co-plots the normalized D/P ratio and the R values (from Eq. 4) against the ^{239}Pu/(^{239}Pu +^{235}U) mass ratio for the eleven assemblies being considered. Both the D/P ratio and the R values show a negative correlation with the ^{239}Pu mass fraction. As the ^{239}Pu fraction increases, the prompt response increases slightly but the delayed response decreases, hence the D/P ratio should decrease with an increasing ^{239}Pu fraction. For the time correlated discrimination, as the ^{239}Pu fraction increases, less of the counts are in the "tail" so R decreases. The normalized D/P ratio exhibits a greater variation over the mass ratio range (1 to ~3), i.e greater sensitivity to the ^{239}Pu fraction variation compared to the normalized R-values (1 to ~1.5).

3. Summary

The R-parameter does in fact capture the shape, or the temporal response of the induced fission response, giving the relative change in the ^{239}Pu mass fraction present in an assembly. A well-characterized sample must be used to calibrate the system in order to measure actual ^{239}Pu mass fractions. Also, the influence of presence of ^{241}Pu is not considered here for the time-correlated discrimination approach. This study is intended to summarize the initial concept of applying the time-correlated neutron signal to discriminate between ^{239}Pu and ^{235}U, however the gate optimization will be unique for different detector configurations. Although the discrimination ratio, R, is not as large as the D/P ratio among the eleven assemblies, it may provide enough information for discrimination without requiring a large, external interrogation source needed by the delayed neutron measurement.

4. Acknowledgements

The authors would like to acknowledge the support of the Next Generation Safeguards Initiative (NGSI), Office of Nonproliferation and International Security (NIS), National Nuclear Security Administration (NNSA).

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LA-UR 11-02592

Combining PNAR-FC and SINRD to Determine Plutonium Content in Spent Nuclear Fuel

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Abstract:

The combination of Passive Neutron Albedo Reactivity with Fission Chamber (PNAR-FC) and Self-Interrogation Neutron Resonance Densitometry (SINRD) to quantify plutonium content of a spent nuclear fuel assembly was modeled using Monte Carlo N-Particle eXtended (MCNPX) simulations. Both of these methods rely on neutrons produced by ^{244}Cm in the spent fuel to self interrogate the content of the spent fuel. PNAR-FC measures the reactivity of the spent fuel assembly, which is a measure of the fissile material inside the assembly of which plutonium is a component. SINRD, on the other hand, uses ^{235}U and possibly but not necessarily ^{239}Pu fission chambers to quantify ^{235}U and ^{239}Pu content in the spent fuel by detecting the change in the neutron flux in different parts of the neutron energy spectrum. Using these two techniques should allow for increased precision and accuracy of the plutonium content inside a spent fuel assembly and increased probability of detection of a fuel pin diversion scenario. This work is funded as part of the Next Generation Safeguards Initiative by the United States Department of Energy and is being executed through several national laboratories and universities inside the United States.

Keywords: spent fuel, plutonium, nuclear safeguards, non-destructive assay

1. Introduction

The effort to safeguard nuclear material inside the nuclear fuel cycle has been ongoing for almost half a century. An area that has always presented problems to nuclear safeguards professionals has been accounting for material inside spent nuclear fuel. Spent nuclear fuel presents a difficult problem due to its unique mixture of fissile isotopes, neutron absorbers, and high radiation levels.

Many techniques have been devised to ensure fissile material in spent nuclear fuel is not diverted. Currently, most techniques rely on relative change across an assortment of spent assemblies to ensure broad diversion has not occurred. However, the actual fissile material inside the assembly is not directly assessed. Therefore, if a method could be devised to measure fissile material directly, it would be of great benefit to the safeguards community.

The Next Generation Safeguards Initiative (NGSI) is tasked with developing methods that will improve upon the currently used techniques. Part of that work has been put into research of new measurement processes that give an indication of fissile material. Two of the techniques researched for this effort at Passive Neutron Albedo Reactivity with Fission Chambers (PNAR-FC) and Self-Interrogation Resonance Densitometry (SINRD). When combined, these 2 techniques offer information from which elemental plutonium mass in a spent fuel assembly can be estimated.

This project is funded by Office of Nuclear Safeguards & Security (NA-241) as part of the National Nuclear Security Administration under the Department of Energy.

2. Spent Fuel Library

As part of the larger effort, a library of 64 assemblies was created for the Monte Carlo N-Particle (MCNP) simulation code. The library consists of 4 different enrichments burned to 4 different burnups and then cooled to 4 different cooling times. Each assembly was designed to be a standard 17x17 PWR assembly, with homogenous fuel and constant enrichment across the assembly. The assemblies were burned using CINDER and MCNP.

3. PNAR-FC

PNAR-FC is a lightweight, low-cost system that is designed to interrogate a spent fuel assembly with thermal neutrons to show the response of three isotopes: ^{235}U, ^{239}Pu, and ^{241}Pu. The basic PNAR method ratios singles neutron counts from the assembly over the singles neutron counts from the assembly surrounded by a cadmium liner. Effectively, this shows the effect that thermal neutrons re-entering the assembly have on the three fissile isotopes of interest. Due to the high fission cross sections for previously mentioned isotopes, the ratio directly measures the mass of those three isotopes combined. Figure 1 (by Conlin) shows the PNAR-FC detector geometry.

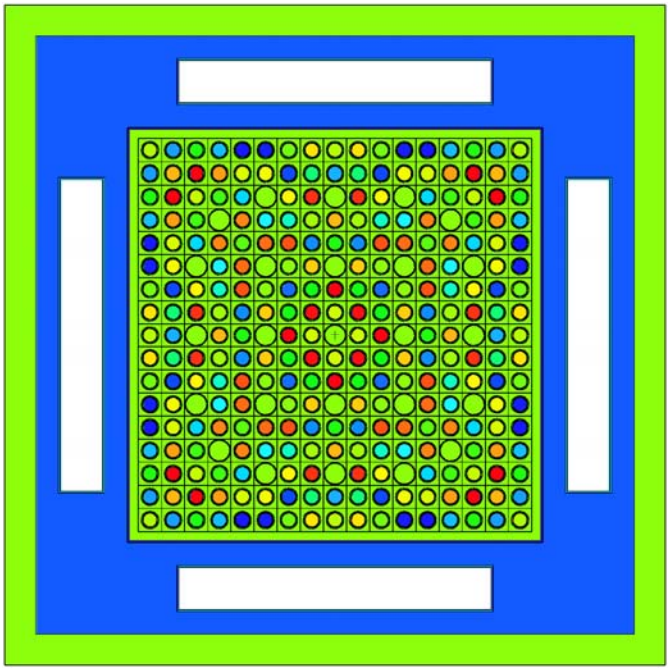


Figure 1: PNAR-FC detector geometry with modeled spent fuel assembly inside. The assembly is surrounded by borated water.

Not seen in Fig.1 is the 1 mm cadmium liner that would surround the fuel assembly for half of the measurement.

In order to quantify our ratio according to the mass of the three isotopes, a $^{239}Pu_{Effective}$ mass has been proposed. The $^{239}Pu_{Effective}$ mass gives the expected contribution of the signal of the three isotopes

through the use of a C_1, C_2 approach. It is similar in concept to the more traditional $^{240}Pu_{Effective}$ mass in safeguards, but it is more tailored to this particular instrument. Equation 1 defines our $^{239}Pu_{Effective}$ mass as

$$^{239}Pu_{Effective} = C_1 M_{U235} + M_{Pu239} + C_2 M_{Pu241} \quad 1.$$

where M_{U235}, M_{Pu239}, M_{Pu241} is the mass of the isotopes ^{235}U, ^{239}Pu, ^{241}Pu respectively and C_1 and C_2 are constants that weight the relative contribution of neutron production from the respective isotope. C_1 and C_2 are given by equations 2 and 3

$$C_1 = \frac{(\bar{\upsilon}_{U235} - 1)\sigma_{f,U235}}{(\bar{\upsilon}_{Pu239} - 1)\sigma_{f,Pu239}} \quad 2.$$

$$C_2 = \frac{(\bar{\upsilon}_{Pu241} - 1)\sigma_{f,Pu241}}{(\bar{\upsilon}_{Pu239} - 1)\sigma_{f,Pu239}} \quad 3.$$

where $\bar{\nu}$ is the mean number of neutrons released per fission for an isotope and σ_f is the microscopic fission neutron cross section. The constants represent the relative net neutron production from ^{235}U and ^{241}Pu to ^{239}Pu.

Figure 2 (by Conlin) shows the results for the 64 spent fuel assemblies for PNAR-FC.

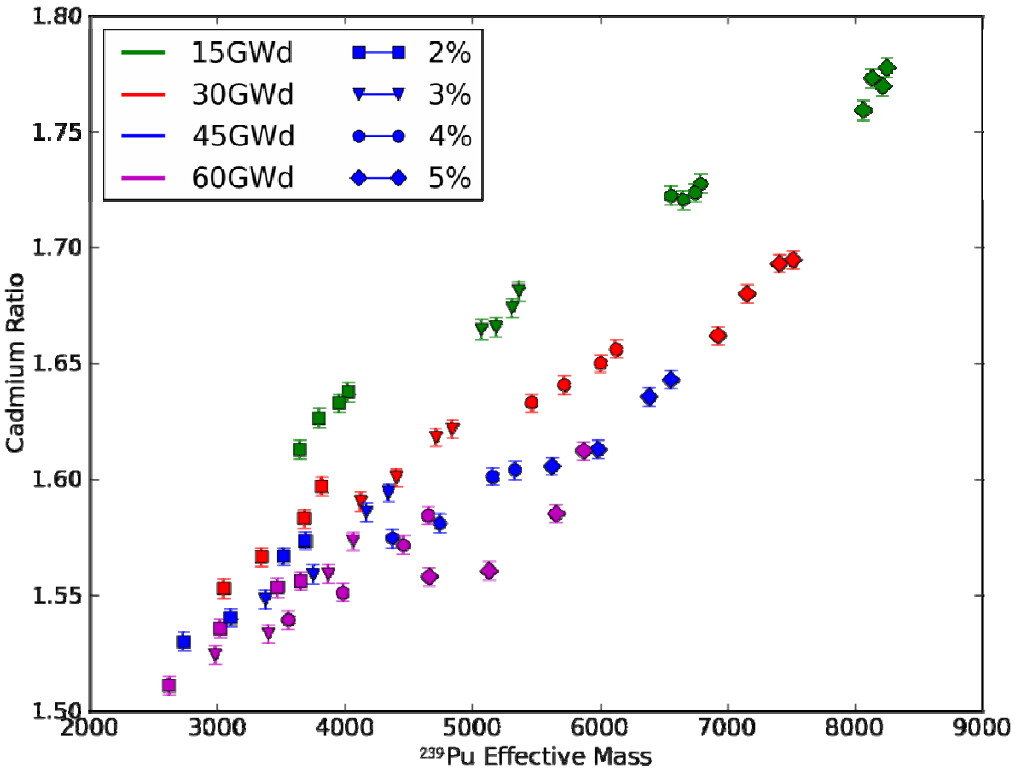
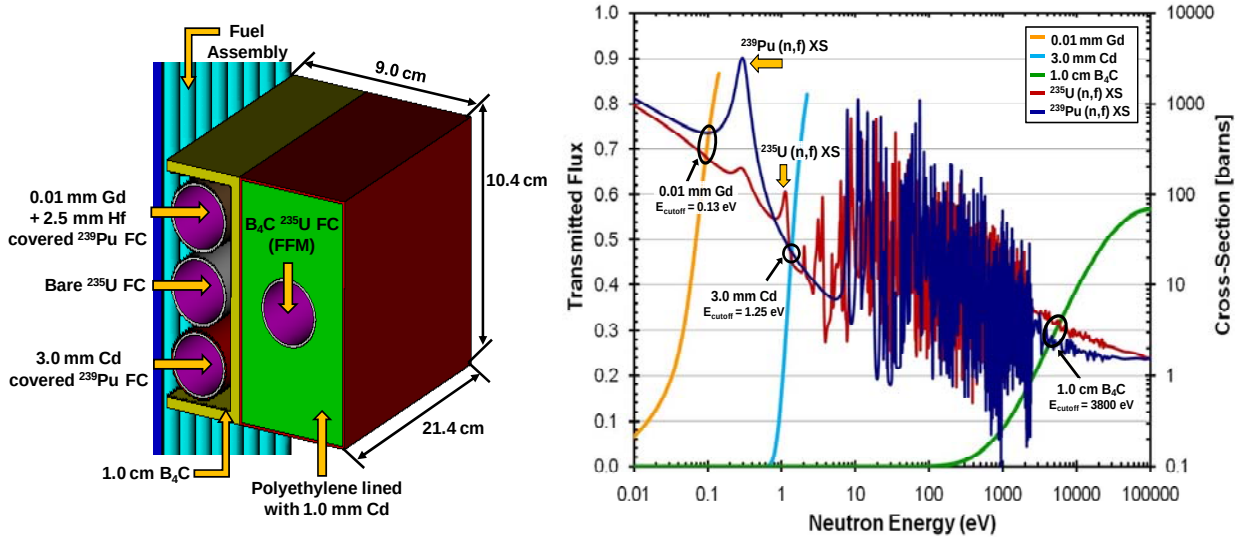


Figure 2: PNAR-FC results for the 64 assemblies in the spent fuel library surrounded by borated water. The cadmium ratio is on the y-axis with $^{239}Pu_{Effective}$ mass along the x-axis.

With Fig. 2, the effect on the cadmium ratio from burnup and initial enrichment can be seen clearly. With an independent measurement of burnup and initial enrichment, a measured cadmium ratio could be related to an effective $^{239}Pu_{Effective}$ mass. That information can be combined with another technique or two to estimate elemental plutonium content in the spent fuel assembly.

4. SINRD

SINRD is another lightweight system that uses the resonance structure of the fission cross sections of ^{239}Pu and ^{235}U to interrogate a spent fuel assembly. SINRD is composed of 4 fission chambers using either ^{235}U or ^{239}Pu as the fissile material. As designed for the 17x17 assembly, SINRD is composed of 3 fission chambers as close to the fuel as possible with one further back surrounded by B_4C and high density polyethylene. 2 of the fission chambers close to the fuel have liners that surround them: one made of Cd and the other a combination of Gd and Hf. The third fission chamber close the fuel assembly is bare. The two fission chambers with liners are ^{239}Pu loaded, while the bare and the poly embedded fission chamber are ^{235}U loaded. Each fission chamber is designed to detect different aspects of the fuel assembly. The bare ^{235}U fission chamber is designed to measure the thermal neutron flux leaving the fuel assembly and the poly embedded ^{235}U fission chamber is designed to measure the fast neutron flux above the resonance energies (FFM for short). The Gd+Hf covered and Cd covered ^{239}Pu fission chambers are designed to measure resonance absorption in ^{239}Pu. The liners can be considered as filters which create a window through which the dominate reactions in the fuel are (n,f) for both ^{235}U and ^{239}Pu. Figure 3 (by LaFleur) shows the design of the detector as well as energy window for the 2 ^{239}Pu fission chambers.



(a) SINRD Detector Configuration **(b) Cut-off Energies of SINRD Absorber Filters**

Figure 3: On the left is the design of the detector. The right shows the (n,f) cross sections for both ^{235}U and ^{239}Pu as well as the window provided by using the different liners.

Fig. 3 shows how SINRD receives its signal. First, the detector has been designed to measure the thermal flux coming from the assembly with the bare fission chamber. Second, two fission chambers are used to create a window around the low energy resonances of ^{235}U and ^{239}Pu through the use of filters. Third, a large portion of the thermal neutron flux is blocked by the B_4C and Cd liner around the

polyethylene which allows the poly-embedded fission chamber to measure the fast flux emitted by the assembly. Using all of these signatures, some interesting correlations can be made.

By taking the ratio of the signal from the FFM to the difference of the Gd+Hf and Cd covered fission chambers, a clear correlation to ^{239}Pu mass can be seen. Figure 4 shows that correlation.

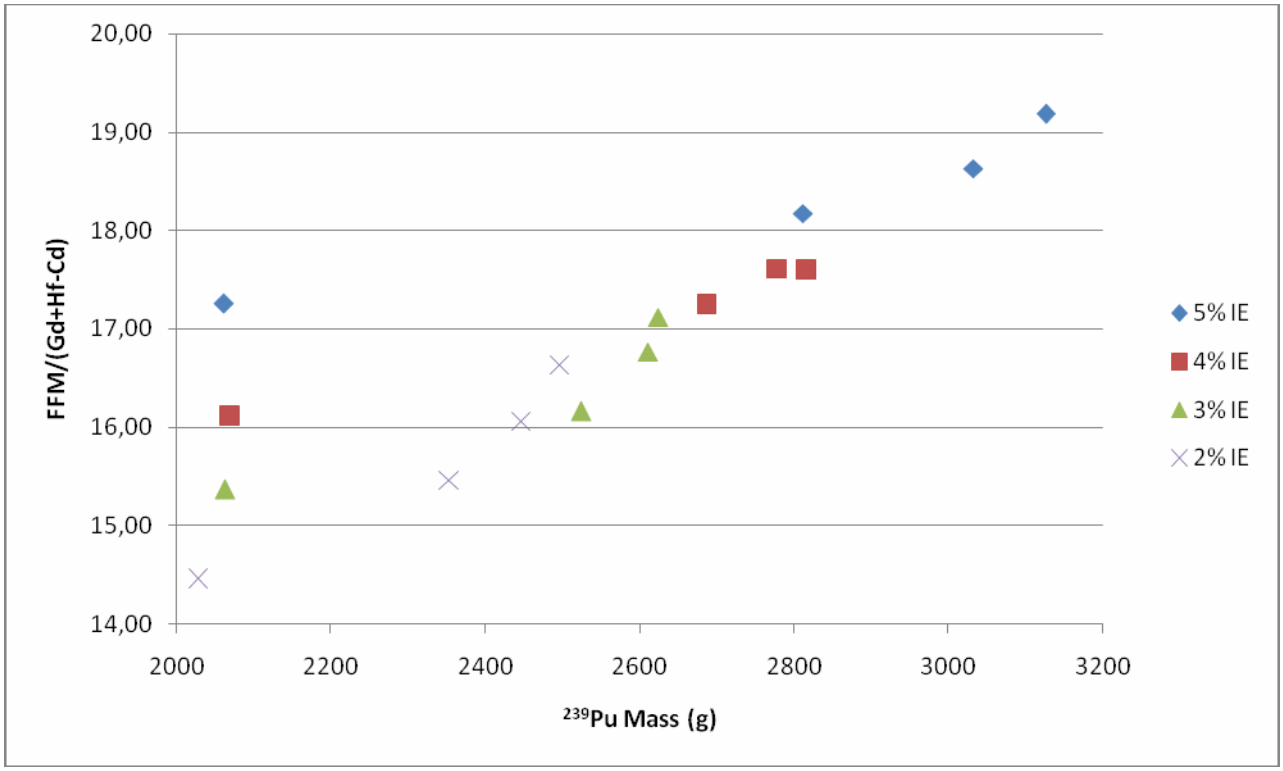


Figure 4: Correlation between FFM/(Gd+Hf-Cd) and ^{239}Pu mass. Shown here are values for 15,30,45,and 60 GWd/MT at 5 years cooled in borated water.

As can be seen in Fig. 4, the correlation between ^{239}Pu mass and the ratio is almost linear for most cases. Later analysis shows that for cases where ^{235}U mass is greater than 3.5 times the ^{239}Pu mass in an assembly this type of analysis is less accurate.

A similar correlation can be made for the ^{235}U. There is a strong correlation between the ratio of the Gd covered ^{239}Pu fission chamber and the bare ^{235}U fission chamber and the mass of ^{235}U. Figure 5 shows that correlation.

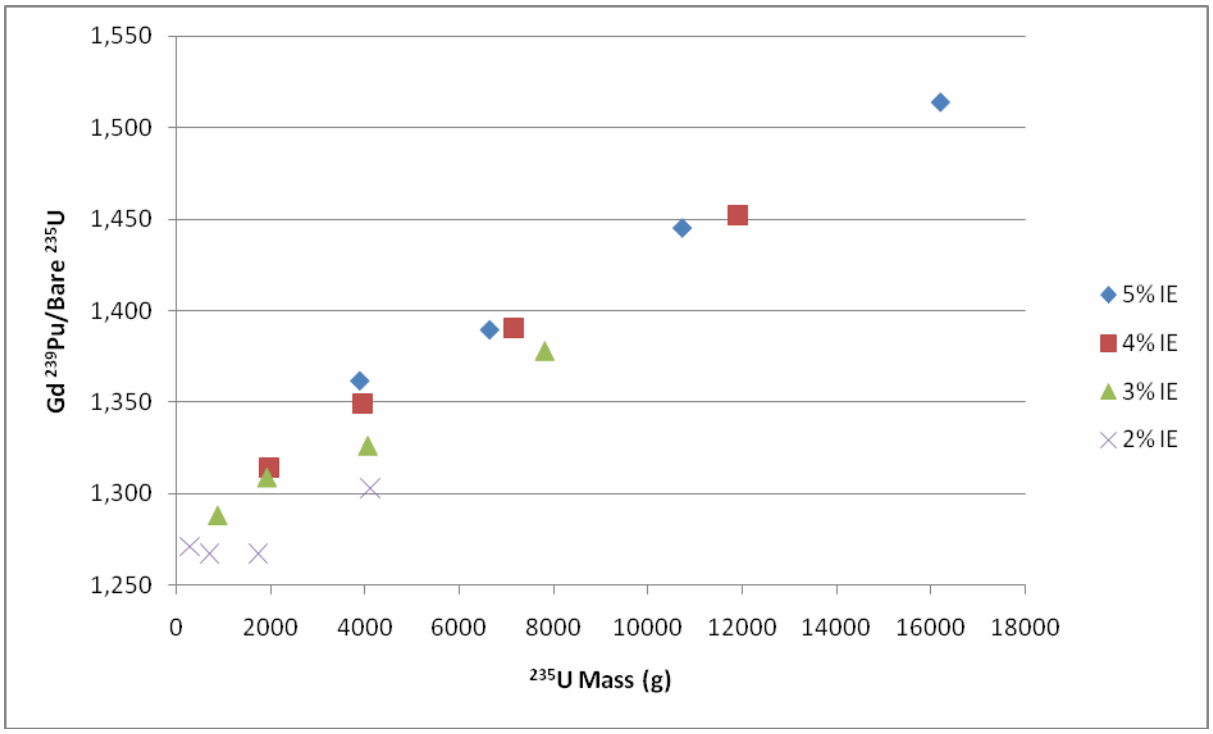


Figure 5: The correlation between the ratio of the Gd covered ^{239}Pu fission chamber and the bare ^{235}U fission chamber and the mass of ^{235}U. Shown here is the data from the 15,30,45,60 GWd/TU burnup cases that are 5 years cooled.

Fig 5 shows the correlation between the ratio of Gd covered fission chamber to the bare fission chamber and the mass of ^{235}U. With a measurement of the initial enrichment of the assembly, most of the ratios have an almost linear response to ^{235}U mass.

It should be noted that this work was done on correlation alone. Additional work is now underway to fully understand what the ratios are actually measuring using new features introduced into MCNPX. Preliminary results have shown that up to 70% of the ratio that indicates ^{239}Pu is actually due to ^{239}Pu. Eventually, a C_1 and C_2 approach will most likely be done.

6. Method to Determine Plutonium Content of Spent Nuclear Fuel

To begin to quantify elemental plutonium in spent nuclear fuel, a few measurements must be made first. The assembly must have a known burnup, initial enrichment, and cooling time. Once those attributes are known, a PNAR-FC cadmium ratio can be made. The cadmium ratio is a measure of the fissile material inside the assembly as shown earlier. This cadmium ratio will give a $^{239}Pu_{Effective}$ mass. In this test case, C_1 and C_2 have already been calculated from the MCNPX simulations for each of the spent fuel assemblies modeled in the spent fuel library. In other cases, an average of the calculated C_1's and C_2's could be used without introducing much uncertainty.

Once the $^{239}Pu_{Effective}$ mass is known, there is an equation with three unknowns: ^{235}U, ^{239}Pu, and ^{241}Pu masses. Once a SINRD measurement has been completed, 2 of those unknowns are known and the third can be solved for. SINRD will provide the mass of ^{235}U and ^{239}Pu, from which the $^{239}Pu_{Effective}$ mass equation can be used to calculate a mass for ^{241}Pu.

At this point, the mass of ^{239}Pu and ^{241}Pu are known. By knowing the burnup and initial enrichment, an estimate of the percentage that ^{239}Pu and ^{241}Pu make up for all plutonium in the assembly can be made using a burnup code. The mass of plutonium in the assembly can then be estimated by dividing the sum of the ^{239}Pu and ^{241}Pu masses by the expected percentage they should make up of all plutonium.

Figure 6 shows the calculated Pu mass in grams as a function of burnup for different initial enrichments at 5 year cooled.

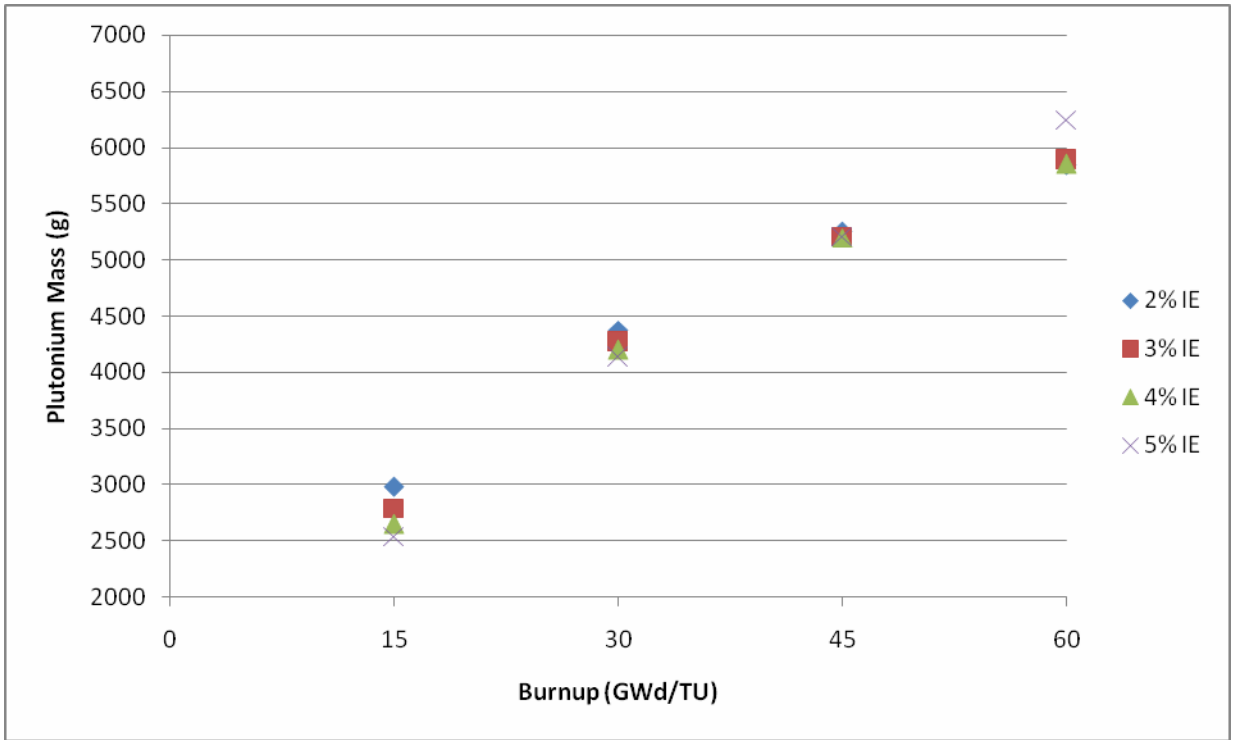


Figure 6: Plutonium mass as a function of Burnup. This mass is calculated using the combination of PNAR-FC data and SINRD data.

As expected, higher initial enrichments produce lower amounts of plutonium at low burnups and higher amounts of plutonium at higher burnups compared to lower initial enrichments. This is to be expected because lower initial enrichments produce more ^{239}Pu at low burnups while they primarily burn ^{235}U which reverses itself at higher burnups. Higher initial enrichments have more ^{235}U to burn at first, and therefore do not burn as much plutonium at higher burnups.

Figure 7 shows the mass of plutonium as a function of burnup at different cooling times for the 4% initial enrichment cases.

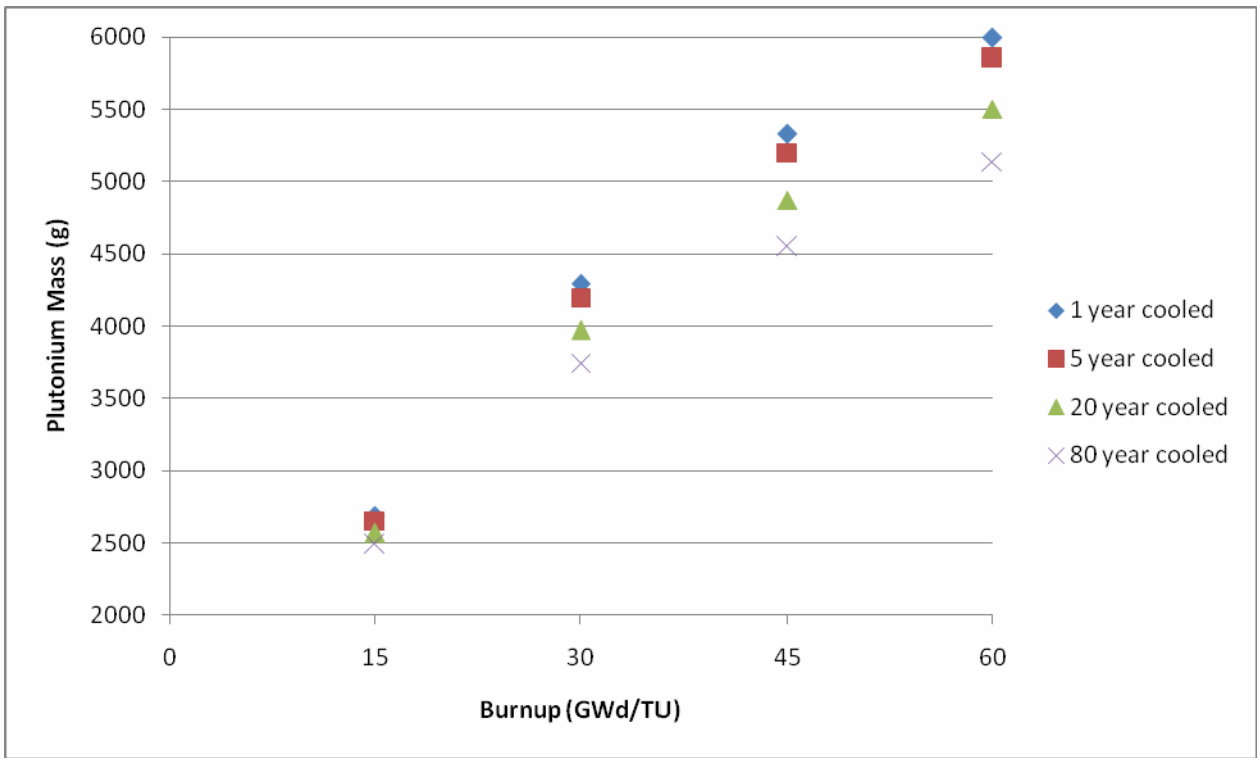


Figure 7: Plutonium mass as a function of burnup for different cooling times for the 4% initial enrichment cases.

Fig. 7 shows how the plutonium mass changes in a 4% initial enrichment assembly as a function of cooling time for different burnups. Again, the pattern falls as it should be expected. Over the time period modeled, only ^{241}Pu and to some extent ^{238}Pu will have a chance to appreciably decay. This change in mass is reflected here. The majority of the lost mass is due to ^{241}Pu decay.

7. Conclusion and Future Work

As has been shown in this paper, it is possible to estimate the plutonium content of a spent fuel assembly using a combination of the PNAR-FC and SINRD detection methods. There are a few caveats for this procedure however. Further analysis must be done on both methods.

Systematic uncertainty needs to be better understood for PNAR-FC. Currently, only the location of the assembly in the detector has been looked at. Other areas of interest are axial burnup distribution and differences in initial enrichment within the assembly.

SINRD needs a better understanding of where the signal in the fission chambers is actually from. All of the work done in this paper is done on correlation. The expected signal in each fission chamber from the fuel assembly is then compared to the known values of ^{235}U and ^{239}Pu. Now, recent research has shown that up to 70% of the detector response is due to the ^{239}Pu fission inside the fuel. A greater understanding of SINRD is needed before this technique could be used in an actual verification regime.

A new library of spent fuel assemblies has already been created, with a more realistic shuffling scheme to better represent actual spent fuel. This library will help benchmark both systems and help to refine their uncertainties.

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2. A.M. LAFLEUR, H.O. MENLOVE, S.J. TOBIN, M.T. SWINHOE. “Development of Self-Interrogation Neutron Resonance Densitometry to Measure the Fissile Content in PWR 17x17 Spent LEU Fuel”, LA-UR 11-00516 (2010)

Neutron Resonance Transmission Analysis (NRTA): Initial Studies of a Method for Assaying Plutonium in Spent Fuel

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Abstract:

Work is underway at Idaho National Laboratory to investigate the Neutron Resonance Transmission Assay (NRTA) technique, assessing its feasibility for quantifying plutonium in spent nuclear fuel (SNF) and for determining the diversion of SNF pins from assemblies. Preliminary results indicate that NRTA has great potential for being able to assay intact SNF assemblies. Relevant in the neutron energy range from 0.1 to 20 eV, it can identify four plutonium isotopes (^{239,240,241,242}Pu), three uranium isotopes (^{235,236,238}U), and six fission products (^{99}Tc, ^{103}Rh, ^{131}Xe, ^{133}Cs, ^{145}Nd, and ^{152}Sm). It can determine the areal density or mass of these isotopes in single- or multiple-pin integral transmission scans. Further, multiple observables exist to allow the detection of material diversion (pin defects) including fast-neutron and x-ray radiography, gross-transmission neutron counting, plutonium resonance absorption analysis, and fission-product resonance absorption analysis. Initial benchmark modelling has shown agreement with previously published experimental data for measurements of individual SNF pins where plutonium assays were experimentally demonstrated to have a precision of better than 3%.

Keywords: spent nuclear fuel, plutonium, assay, neutron, resonance

1. Introduction

New analytical methods are needed to assay spent nuclear fuel to determine fissile material inventories. Ideally, these measurements will be able to assay whole, unaltered commercial-nuclear-reactor assemblies without the need for disassembly, sectioning, or chopping, providing “head-end” inventory data for materials entering current-generation and future spent-fuel reprocessing/recycling facilities. Quantitative measurements of plutonium entries into safeguarded fuel reprocessing facilities prior to material decomposition will provide a higher level of confidence for the actual plutonium mass than found with the current approach, which relies on vendor-supplied burn-up calculations. One potential method to accomplish this task is neutron resonance transmission analysis, NRTA. This paper summarizes recent research at Idaho National Laboratory to begin evaluating NRTA as a method for assaying plutonium in spent nuclear fuel (SNF).[1] This work is part of the U.S. Next-Generation Safeguards Initiative and its goal is to develop a method to assay SNF with a precision of 1% or better.

The NRTA concept is based on solid theoretical principles, has been demonstrated experimentally at the bench scale using commercial spent fuel, and has achieved a plutonium assay measurement precision of 2-4% in ad-hoc testing.[2-4] The technique uses a pulsed accelerator to produce an intense, short pulse of neutrons. These neutrons, travelling at different speeds according to their energy in a time-of-flight (TOF) configuration, are used to interrogate a spent fuel assembly. Neutron transmission through the assembly is monitored as a function of neutron energy (time after the pulse), similar to the way neutron cross-section data is often collected. Neutron detection is performed using a high-rate sensor, such as a lithiated-glass scintillation detector.[3-5] Results are read from the detector as count rate versus time, with faster (higher-energy) neutrons arriving first and slower (lower-energy) neutrons arriving later. The low-energy elastic scattering and absorption resonances of plutonium and other isotopes modulate the transmitted neutron spectrum. The plutonium content in the fuel can be determined by analyzing this attenuation.

The NRTA concept is not new. The first paper (reference 6) on the subject was published in 1975, followed by substantial research through the mid-1980s. Much of the published research literature is from the U.S. National Bureau of Standards (NBS), focusing on applications related to neutron radiography. In particular, the measurement of uranium in waste materials and the assay of isotopic heterogeneities in welds, brazes, and metals has been discussed.[7-12] One NRTA study looked at optimizing the accelerator neutron output.[13] Before NRTA research and publishing literature diminished, around 1984, several papers documented the use of NRTA to characterize a PWR spent fuel pellet, a pellet with approximately 25 GWD/MTU burnup.[2-4] These NRTA spent-fuel papers have provided confirmation that the NRTA concept is a viable and potentially precise spent assay technique.

2. General Overview

The NRTA technique uses low-energy neutrons as the probing radiation, in the 0.1-40 eV energy range. This neutron energy range is at the bottom end of the actinide-resonance range, where most actinides have at least one or more resonances. For the spent fuel actinides the resonances are typically large in magnitude, narrow in width, and fortuitously well-separated resulting in distinctive resonance transmission spectra. Of the hundreds of fission-product isotopes found in spent fuel, only a half-dozen or so have significant resonance structure in the 0.1-40 eV energy range. These fission products have only one or two resonances in this energy range, none interfere with the actinide resonances. Other isotopes present in a SNF assembly – for example, oxygen in the UO_2 and zirconium, tin, iron, chromium, oxygen, niobium, nickel, carbon, and silicon in the Zircaloy-4 clad – also have no resonance structure in the 0.1-40 eV range. The same is true for the hydrogen impurity (hydration) in the clad and air isotopes.

An important part of an NRTA system is the neutron source. The pulsed neutron field needed for NRTA may be produced many different ways; accelerator-based production reactions are perhaps the most convenient. Light-ion reactions with ^{7}Li(p,n) or ^{9}Be(p,n) provide one attractive approach for generating neutrons for NRTA. Prior work has shown bremsstrahlung-based photoneutron sources using electron accelerators operating from 10 to 100 MeV can serve as excellent neutron sources for NRTA. Regardless of the primary source of neutrons, accelerator-based NRTA neutron sources require a neutron moderator assembly to convert high energy neutron to lower energies. High-energy neutrons pass through a low-Z neutron moderator such as polyethylene, light water, or heavy water where they scatter and lose energy. This scattering process yields a continuous distribution of neutrons, varying in energy from the starting energy down to approximately 0.025 eV. It is the portion of the continuum having thermal and epithermal neutrons from 0.1 to 40 eV that is of interest to the NRTA technique.

The thermalized neutrons can then be collimated to create a neutron beam that is directed at a spent fuel assembly. A fraction of the neutrons incident on the fuel assembly scatter out of the beam as they interact with individual fuels pins in the assembly through low-energy elastic scattering, neutron-capture absorption, and neutron-capture fission. The rest of the neutrons pass through the spent fuel assembly unaffected, as the transmitted signal. It is these transmitted neutrons – the modulated beam – that is measured in the NRTA technique. The modulated beam, or transmitted signal, is an integral scan of rows of pins in the assembly and is analogous to how a traditional x-ray radiograph is used to detect the presence of dense objects in the human body (bones) and the location of low density regions (bone cracks). The NRTA technique does not assay individual pins in a SNF, with the exception of the four corner pins.

A schematic representation of an NRTA measurement is shown in Figure 1. The major components include: (1) pulsed particle accelerator, (2) neutron moderator to thermalize neutrons, (3) a filter to remove very low-energy neutrons (below 0.1 eV), (4) time-of-flight drift tube, (5) vertical chamber to contain and cool a spent fuel assembly, (6) mechanism to lift, rotate, and transversely move the assembly, and (7) neutron detectors with associated TOF electronics for data acquisition.

Low-energy fission, capture, and elastic scattering resonances of plutonium and other isotopes modulate the transmitted-neutron spectrum. Spectrum modulation, or neutron attenuation, appears as depressions in the collected data that correspond to energy-specific resonances and, hence, specific

and identifiable isotopes. The plutonium content in the fuel can be determined by analyzing the appropriate depression signals (magnitude or area of the depression).

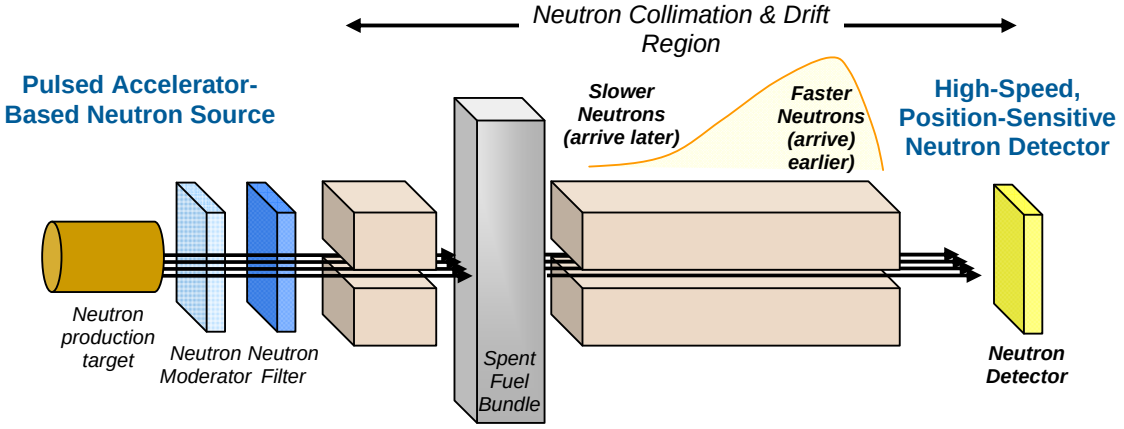

Figure 1: Schematic representation of the NRTA measurement approach.

3. Numerical Simulations

Several MCNP (Monte Carle n-Particle transport) code models have been developed for NRTA feasibility studies, they were all relatively simple in geometry.[14,15] The simplest geometry models included a directed neutron source, a single fuel pin or line of pins, an evacuated flight tube, and flux-tally detector cells. More sophisticated models included a complete pressurized water reactor (PWR) 17 × 17 pin SNF assembly, air-filled drift tubes, and a wide-area neutron beam; the SNF assembly models were taken directly from models developed at Los Alamos National Laboratory that were previously developed as part of the U.S. Next Generation Safeguards Initiative.[16] The PWR spent-fuel pin geometry included a 0.82 cm UO_2 pellet diameter, 0.95 cm diameter clad (no gap), and a fuel pin pitch of 1.26 cm.

In all of the MCNP models the neutron source was uniformly sampled over the desired transmission neutron-energy range. In an actual physical NRTA system a slowing-down neutron source would be expected to exhibit some non-uniformity, in particular, slightly higher flux at higher energies. In the 0-40 eV energy range, with the use of a low-Z neutron moderator with low absorption, a relatively flat and uniform energy-flux distribution would be expected. Also, the benchmark data does not exhibit a significant flux tilt to higher energies. The MCNP models also used two different beam geometries: cylindrical beam and directed point-source beam. The cylindrical beam typically had a diameter less than the UO_2 pellet diameter (<0.82 cm) and was directed through the fuel pin center, such that no neutrons struck the detector flux tally cell without first passing through the UO_2 pellet. The cylindrical-beam radius was also varied for some studies. The directed point source is essentially a cylindrical beam with zero radius: a point source directed in a particular direction creating a line of neutrons. The directed point source is useful in order to assess the effect of pellet curvature on the transmission signal.

Evaluated Nuclear Data File VII (ENDF-7) was used for most of the numerical simulations.[17] ENDF-5 and ENDF-6 data were used initially and results calculated with these data compare well with the ENDF-7 data.

3.1 The Benchmark Problem

In order to develop a level of confidence in the MCNP codes, models, and cross section data, it seemed prudent to compare a calculated transmission spectrum against an actual measured transmission spectrum. Two measured spectra were available in the published literature for ad-hoc experiments using a single spent fuel pin.[3] From the cited reference, the two measured spectra were based on UO_2 samples cut from a single spent fuel pin, one from the center of the pin and the other from the end of the pin. Both cylindrical samples were approximately 1.0 cm in diameter and 2.5

cm in length (probably a PWR fuel pin). The fuel pin had an estimated burnup of approximately 25 GWD/MTU. No initial enrichment or cooling time was given, but prior experience indicates that a PWR assembly with approximately 25 GWD/MTU back in the 1970s timeframe was probably around 3.2 wt% ^{235}U.

For comparison purposes, a spent fuel assembly from the NGSI SNF library with burnup characteristics closest to these samples was chosen for the transmission calculation. The closest match used a 3.0 wt% ^{235}U initial enrichment, 30 GWD/MTU burnup, and a 5-year cooling time. A material composition (m704) was arbitrarily chosen from a peripheral pin in the NGSI assembly for the MCNP transmission calculation. The burnup characteristics between the actual measured spent-fuel composition and the NGSI composition are not exact but are reasonably close.

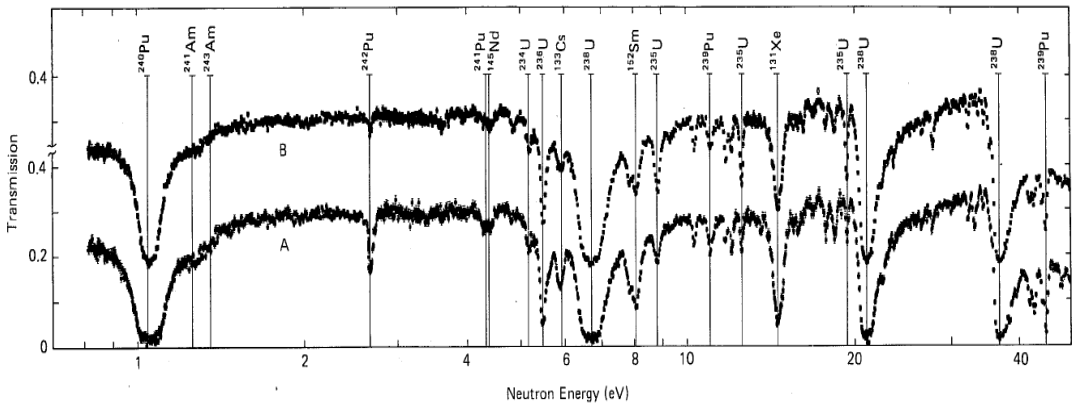

Figure 2: Measured resonance transmission spectra as a function of neutron energy for the NBS spent nuclear fuel samples (Spectrum A is for fuel cut from the center of the fuel pin and spectrum B is cut from one end of the fuel pin).

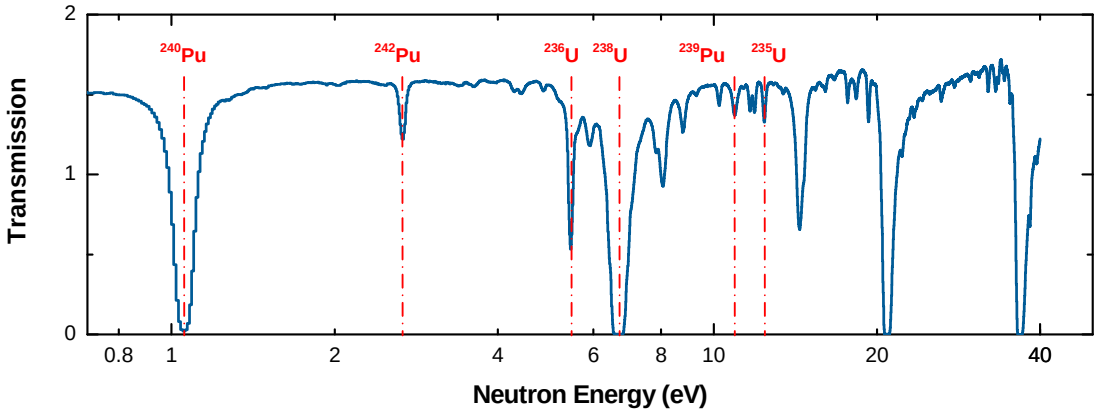

Figure 3: MCNP-calculated transmission spectrum as a function of neutron energy through a single spent fuel pin; important isotopes have been highlighted in red.

Figure 2 shows the two measured transmission spectra from the reference paper. Spectrum A , the lower spectrum, is for the sample cut from the center of the fuel pin, and spectrum B, the upper spectrum, is for the sample taken from the end of the fuel pin. Note the ordinate axis discontinuity in this figure. The resonance depressions correspond to specific actinide and fission product isotopes and are clearly marked in the figure. It is interesting to note the deeper depression in spectrum A for the ^{242}Pu resonance at 2.65 eV. This is due presumably to the higher burnup in the middle of the SNF pin, and correspondingly higher ^{242}Pu concentration. Also, the ^{235}U depression at 8.8 eV is deeper for spectrum B relative to A, due to the higher ^{235}U concentration remaining at the end of the pin relative to the center (due to the lower burnup at the end of the SNF pin).

Figure 3 shows the corresponding MCNP-calculated transmission spectrum. In the NGSI SNF library the spent fuel composition is assumed to be axially uniform, hence the calculated spectrum represents an average over the length of the fuel pin; one might expect the magnitude of the resonance depressions to lie between the two measured spectra. For comparison purposes, the MCNP-calculated spectrum was arbitrarily normalized to the potential scattering levels of spectrum A and B which appear to be approximately equal.

3.2 The PWR SNF-Assembly

A PWR fuel assembly was chosen as the basis for our initial analyses of the NRTA technique. This particular fuel assembly design is a 17 × 17 array with 264 fuel pins and 25 guide tubes. The fuel pins and guide tubes are symmetrically arranged such that each assembly quadrant is a mirror-image of an adjacent quadrant. For NRTA calculations the 25 guide tubes are assumed to be voided or air-filled; the central pin location in the assembly is a guide tube. Fuel pins consist of a UO_2 pellet (0.82 cm diameter) and a Zircaloy-4 clad tube (0.95 cm diameter). The Zircaloy-4 guide tube was assumed to be 1.142 cm in diameter with a wall thickness of 0.042 cm. The pin pitch was 1.26 cm and the overall 17 × 17 array had the dimensions of 21.42 × 21.42 cm. The fuel rods had a length 365.76 cm (12 ft).

Figure 4 shows a cross sectional view of PWR 17 × 17 fuel assembly. The 25 white circles in the assembly are the guide tubes (air-filled), the remaining colored circles represent the fuel pins. Nine (9) vertical lines are drawn over the left half of the assembly and the center row (row 9). In addition, 17 diagonal lines are drawn over the lower half the assembly. At the end of each line is the number of fuel pins in that line. Note that the guide tubes are not counted in the line totals. The pin totals in this figure range from 1–17. Of the 17 possible vertical lines (only 9 shown in the figure), 3 lines would have 12 pins, 2 lines 14 and 15, and 10 lines 17. The 3 lines with 12 pins are noteworthy because they are centrally-located, each line containing 5 guide tubes. For the 17 diagonal lines, only 3 lines have more than 12 pins. It can be shown for the PWR 17 × 17 assembly that, if the NRTA technique can assay 12 pins in a row in a single integral transmission measurement, then every pin in the assembly can be part of one or more integral transmission measurements; complete assay coverage of all the assembly pins would then be possible.

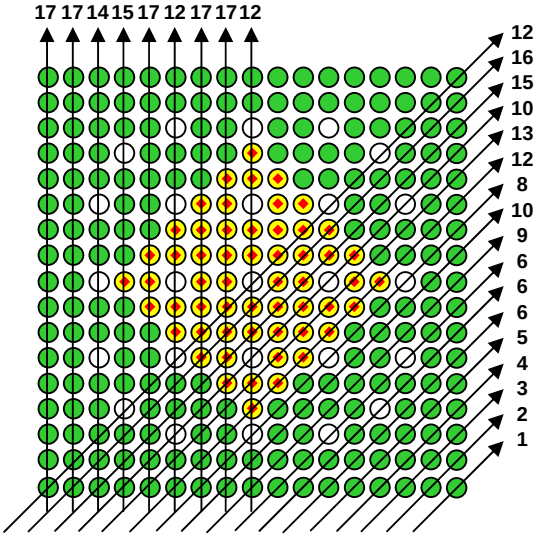

Figure 4: The number of consecutive pins in a row for different views through a 17 x 17 PWR fuel assembly.

3.3 Conceptual Studies

Fourteen conceptual studies have been carried out to examine different aspects of the NRTA measurement technique. The results of these studies are fully documented in reference 1. The primary goal of the studies was to better understand the basic physics behind the NRTA technique and to preliminarily assess its ability to assay a PWR spent fuel assembly. Parametric variables

considered in these studies included spent-fuel isotopics, burnup, cooling time, background neutron emissions, and characteristics of the collimated neutron beam. The results of these conceptual studies provide a preliminary basis upon which a conceptual NRTA system design has begun to emerge. It must be stated that the conceptual studies are not complete. Additional conceptual studies are expected to continue into the future, with the possibility that some of the conceptual studies here will be expanded or re-worked as needed. The fourteen parametric studies are listed in Table 1.

1. Fresh Fuel Performance
2. Differences Between Fresh Fuel & Spent Fuel
3. Effects of Burnup
4. Effects of Cooling Time
5. Effects of Neutron Beam Diameter and Scattering
6. Performance of a Fan-Shaped Irradiation Beam
7. Performance of an Ideal Pencil-Beam Irradiation
8. Transmission Through Multiple Fuel Pins vs. One Pin
9. Differences Between Evacuated and Air-Filled Drift Tubes
10. Potential Interferences from ^{103}Rh at 0.3 eV
11. Inherent SNF Neutron Emission & Impact on Signal Strength
12. Inherent SNF Photon Emission & Impact on Signal Strength
13. Feasibility of Detecting Fuel-Pin Diversion
14. Feasibility of Detecting Missing Fuel Pins

Table 1: Fourteen conceptual study topics for an initial analysis of the NRTA technique.

Studies 1 and 2 demonstrated the driving mechanisms leading to shape in the NRTA spectra. Oxygen and the fuel-clad materials do not play a major role in the spectra. Studies 3 and 4 showed an expected relationship between resonant absorption and the removal of neutrons in the NRTA TOF spectra, scaling with the concentration of the actinides and fission products in the fuel. Studies 5 through 7 were carried out to understand how neutron scattering might serve to blur the resonant features of NRTA spectra. The impact is minimal.

Study 8 investigated the change in transmitted signal versus the number of pins. The results of this study are shown in Figure 5. Transmission through one pin reduces the neutron intensity to ~70% of the untenanted intensity (no pins). Transmission through eight pins produces a neutron intensity roughly 6% of the value for the case with one pin.

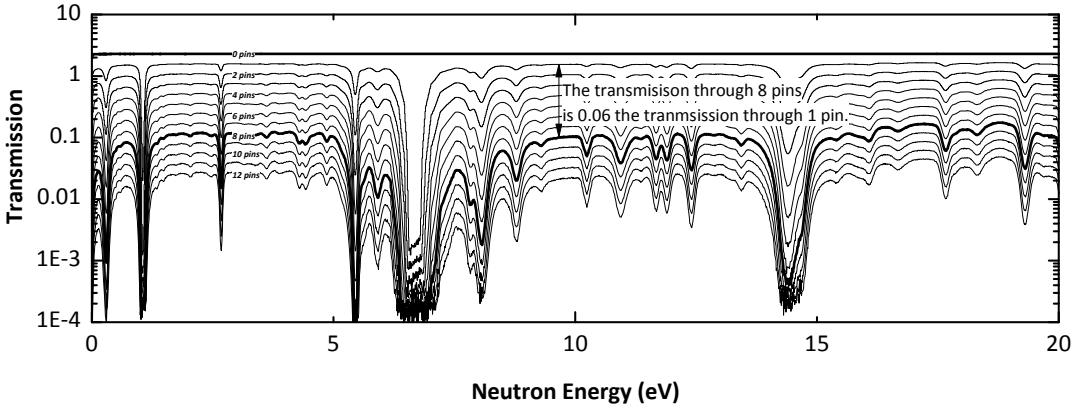

Figure 5: MCNP-calculated transmission spectrum as a function of neutron energy through multiple pins (study 8 from Table 1).

Study 9 indicates the value of using an evacuated flight tube versus and air filled tube. Flight tubes of 1, 2, and 4 meters result in attenuation losses of 4, 8, and 16% compared with evacuated flight tubes of the same length. Study 10 examined if the fission product ^{103}Rh, with a resonance at 1.25 eV, would pose a significant interference for measuring the 1.07-eV resonance of ^{240}Pu. For ^{103}Rh concentrations found in fuel ranging from 0-60 GWD/MTU no significant interference was observed. Studies 11 and 12 evaluated the background signals present with SNF and their potential impact on NRTA measurements. Problems caused by these neutron and photon emissions from SNF can minimized by keeping the fuel 3 or more meters away from the detector system and through the judicious use of shielding. Studies 13 and 14 evaluated how well NRTA can detect the diversion of a single fuel pin anywhere in the bundle (replaced with an unirradiated fuel pin) and removal of a fuel pin (no replacement). The NRTA technique is particularly robust in being able to detect both of these scenarios. Both diversion cases are readily detected using fast neutron radiography alone, thermal neutron radiography, and resonant analysis of the fission products ^{131}Xe or ^{152}Sm.

4. Summary

A set of fourteen conceptual studies have been carried out as part of an initial evaluation of the Neutron Resonance Transmission Assay technique for assaying spent nuclear fuel.[1] Results from the simulations suggest sufficient transmission strength and signal differentiability to assay a minimum of 8 pins in a row, more pins may be possible depending upon the intensity of the neutron source used for the measurements. For an 8-pin maximum, 64% of the fuel pins in an assembly can be part of an integral transmission assay measurement. Collimated neutron beams or neutron detector designs that can spatially resolve a fuel-pin diameter have the potential to assay up to 12-pin rows. For a 12-pin maximum, 100% of the fuel pins in an assembly can be part of one or more integral transmission assay measurements. Prior work has shown that a measurement precision of 3% or better can be achieved for assaying a single fuel pin using an ad-hoc experimental arrangement. Continued work is underway at INL to quantity the neurons source strength needed to be able to reach a minimum assay precision of 1% for transmission through 8 pins.

A summary of some of the strengths of the NRTA technique, as identified from these scoping studies, is presented below.

1. NRTA has the potential for accurate assay measurements with a precision in the range of 1% - 4% uncertainty.
2. NRTA produces distinctive resonance-transmission spectra that can uniquely identify specific actinide and fission product isotopes. The method detects and measures plutonium isotopes directly; it does not rely on correlations or the effective ^{240}Pu concept.
3. In addition to ^{239}Pu, NRTA can identify and assay several additional important fissionable isotopes and spent-fuel actinides directly, including ^{235}U, ^{236}U, ^{238}U, ^{240}Pu, ^{241}Pu, and ^{242}Pu.
4. NRTA can identify the presence of ^{234}U, ^{241}Am, and ^{243}Am. Americium-241 is of particular relevance in higher burn-up fuels (45-60 GWD/MTU) with large cooling times (>5 years).
5. NRTA does not suffer resonance interference effects between ^{235}U, ^{236}U, ^{238}U, ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{242}Pu; the resonances are strong, narrow, and well-separated.
6. NRTA can identify 6 resonant fission-product isotopes (^{99}Tc, ^{103}Rh, ^{131}Xe, ^{133}Cs, ^{145}Nd, and ^{152}Sm) which can potentially be used to estimate assembly burnup, cooling time, and diversion, and to verify operator-reported burn-up values.
7. The neutron resonance transmission analysis technique is a mature technology with a solid foundation in theoretical physics.
8. Nuclear data for spent fuel actinide and fission product isotopes are known to high accuracy, especially the total cross section and resonance parameter data needed for assay determinations.
9. NRTA system calibrations with pin/assembly standards can be straightforwardly used to reduce NRTA systematic errors.
10. An NRTA system can be designed to be insensitive to spent-fuel gamma radiation.

11. If a photoneutron source is used as the NRTA radiation source, the accelerator's gamma flash precedes the NRTA measurement without interference and may be useful for radiography.
12. NRTA does not suffer resonance interferences from the presence of oxides in fuel. (Oxides do, however, attenuate the transmitted signal to a small extent.)
13. NRTA does not suffer resonance interferences from zircaloy cladding; only minor signal attenuation occurs due to the cladding because of the relatively small zirconium total-neutron-attenuation cross section.
14. NRTA does not suffer resonance interference from hydrogen impurities in the cladding.
15. The NRTA signal loss from the presence of hydrogen in the cladding is negligible.
16. NRTA does not suffer resonance interferences from the multitude of spent-fuel fission products. The single resonances from each of the six resonant fission products identified so far that are in the energy range of interest for NRTA (i.e., ^{99}Tc, ^{103}Rh, ^{131}Xe, ^{133}Cs, ^{145}Nd, and ^{152}Sm) do not interfere with the actinide-isotope resonances.
17. The multiplication factor for a spent fuel assembly in vacuum or air is small; multiplication neutrons are therefore relatively minor and do not significantly affect the transmitted signal for NRTA measurements.
18. Background neutrons from spent fuel (spontaneous fission and alpha-n) should be a minor impact through the judicious use of neutron shielding and since there will be a small solid angle to detectors. Background signal subtraction may also be used.
19. No exotic-isotopic, high-purity threshold detectors are required; NRTA neutron detectors are total count detectors, and detection can be performed using standard technology.
20. Vacuum in the NRTA flight tube may not be required, especially for system designs employing relatively short flight tubes (2-4-meter); in these cases, flight tubes could be air-filled without serious signal attenuation.
21. The NRTA approach may be accurately modelled using conventional neutronics tools, such as MCNP. Relatively simple MCNP models are adequate for NRTA simulations.
22. The NRTA system is suitable for incorporating other NGSI fuel-assay techniques.

A summary of some of measurement requirements for the NRTA technique, as identified from these scoping studies, is presented below.

1. The temperature of the spent fuel assembly (spent fuel assembly decay heat) may need to be measured/controlled to eliminate the impact of Doppler broadening on the transmitted spectra.
2. The fuel to be assayed may not be immersed in water.
3. The range of penetrability of low-energy neutrons into a PWR assembly is fundamentally limited; multiple orthogonal views must be used to reconstruct an assembly's complete burn-up and Pu-inventory profile.
4. An intense thermal/epithermal pulsed neutron source is needed.
5. A large physical infrastructure is needed to accommodate the accelerator, time-of-flight tube, detectors, and data analysis system. (However, these requirements will likely be small in comparison with other physical infrastructure requirements needed to allow for the handling of spent fuel.)
6. New data analysis tools are needed.

5. Acknowledgements

The authors would like to acknowledge and thank Dr. Steve Tobin of Los Alamos National Laboratory for his assistance in coordinating and explaining the overarching goals of the NGSI spent fuel assay project, and for allowing us access to resources developed at his laboratory to support spent-fuel assay research and development. The work in this report was sponsored by the U.S. Department of

Energy's Next Generation Safeguards Initiative (NGSI), Office of Nonproliferation and International Security (NIS), National Nuclear Security Administration (NNSA).

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Comprehensive modelling of the hybrid K-edge/K-XRF measurements of actinide solutions: development and experimental validation of a dedicated software program

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Abstract:

The combination of the K-edge transmission measurements with the energy dispersive X-ray fluorescence analysis (XRF), known as the hybrid K-edge/K-XRF densitometry (HKED), has become an indispensable utensil in the nuclear safeguards analytical toolbox that is applied at large-scale spent nuclear fuel reprocessing facilities on a routine basis for measuring actinide concentrations in technological solutions for nuclear material accountancy and process control purposes. Previously we have reported on the development of a Monte Carlo based modelling approach for predicting the XRF response of HKED instruments in terms of the X-ray peak intensities. In the current work the approach has been further extended to include the full XRF spectrum modelling. The simulation module developed has been finally integrated into a dedicated user-friendly software program. It is intended for the support of the calibration and spectrum analysis of the HKED-XRF measurements of actinide solutions from the conventional reprocessing as well as "non-standard" actinide mixtures and sample matrices from the R&D on the advanced reprocessing technology. The paper presents the newly developed software program together with the results of its experimental validation performed with the use of a variety of samples. These include U/Pu solutions encountered in the PUREX process, the U/Pu solutions with heavy metal matrices arising from pyrochemical processing, as well as the Th and mixed U-Th nitric acid solutions.

Keywords: hybrid densitometry; X-ray fluorescence analysis; Monte-Carlo modelling; spectrum; actinides

1. Introduction

The combination of the K-edge transmission measurements with the energy dispersive X-ray fluorescence analysis (XRF), known as the hybrid K-edge/K-XRF densitometry (HKED), has become an indispensable utensil in the nuclear safeguards analytical toolbox that is applied at large-scale spent nuclear fuel reprocessing facilities on a routine basis for measuring actinide concentrations in technological solutions for nuclear material accountancy and process control purposes [1]. Previously we have reported on the development of a Monte Carlo based modelling approach for accurate predicting the XRF response of HKED instruments in terms of the X-ray peak intensities [2]. In the current work the approach has been further extended to include the full X/γ-ray spectrum modelling for all measurement channels of an HKED instrument

including the transmission, X-ray fluorescence and passive gamma. The simulation module developed has been finally integrated into a dedicated user-friendly HKED Response Generation Program HKED-RGP. In this paper we describe the program and present the first results of its experimental validation that by now has been done for the XRF fluorescence channel of an HKED instrument operated at the Institute for Transuranium Elements (ITU), Karlsruhe.

2. HKED-RGP overview

2.1. General description

The program architecture is schematically presented in Fig. 1. It includes the following main components:

- a multi-tabbed intuitive interface (see screenshots in Figs. 2-4) that guides a user through different steps of a simulation process, such as the definition of sample properties, the creation of an excitation source, the physical spectrum modelling, the detector response characterization, setting up of the electronics parameters for data acquisition, and the detector spectrum simulation;

- a multiple table relational database for storing: (i) the pre-defined geometries of HKED instruments, (ii) the user-defined data, such as the analyzed sample and excitation source properties, the detector configurations and electronics parameters, (iii) the calculation parameters and results of calculations, e.g. the energy distributions of excitation sources, the physical spectra of photons emitted/scattered by a sample, the detector responses and simulated photon spectra, (iv) the physical data including the properties of isotopes, isotope mixtures, chemical elements and compounds, as well as a comprehensive array of photo-atomic data;

- a number of dynamic-link libraries modules for performing specific calculations, such as the modelling of the excitation X-ray spectrum, the detector response characterization, the modelling of a physical spectrum of photons emitted/scattered by an analyzed sample, and the generation of a resulting detector spectrum.

The basic feature inherent to the program's architecture and implementation approach allows the adaptation to an arbitrary HKED instrument. The program offers the capability to monitor the simulation results at each modelling step, either in graphical or numerical form and to save the results in a database for future use. In this way, for instance, it is easy to generate spectra for different instruments and measurement channels of the same instrument, as well as for different combinations of the user-defined samples, excitation sources, detectors and electronics configurations.

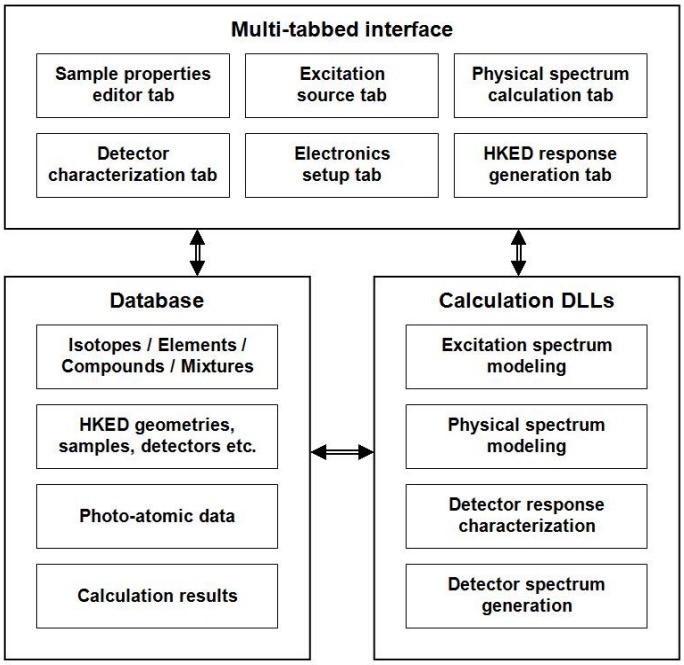

Fig. 1. A schematic diagram representing the HKED-RGP program's architecture.

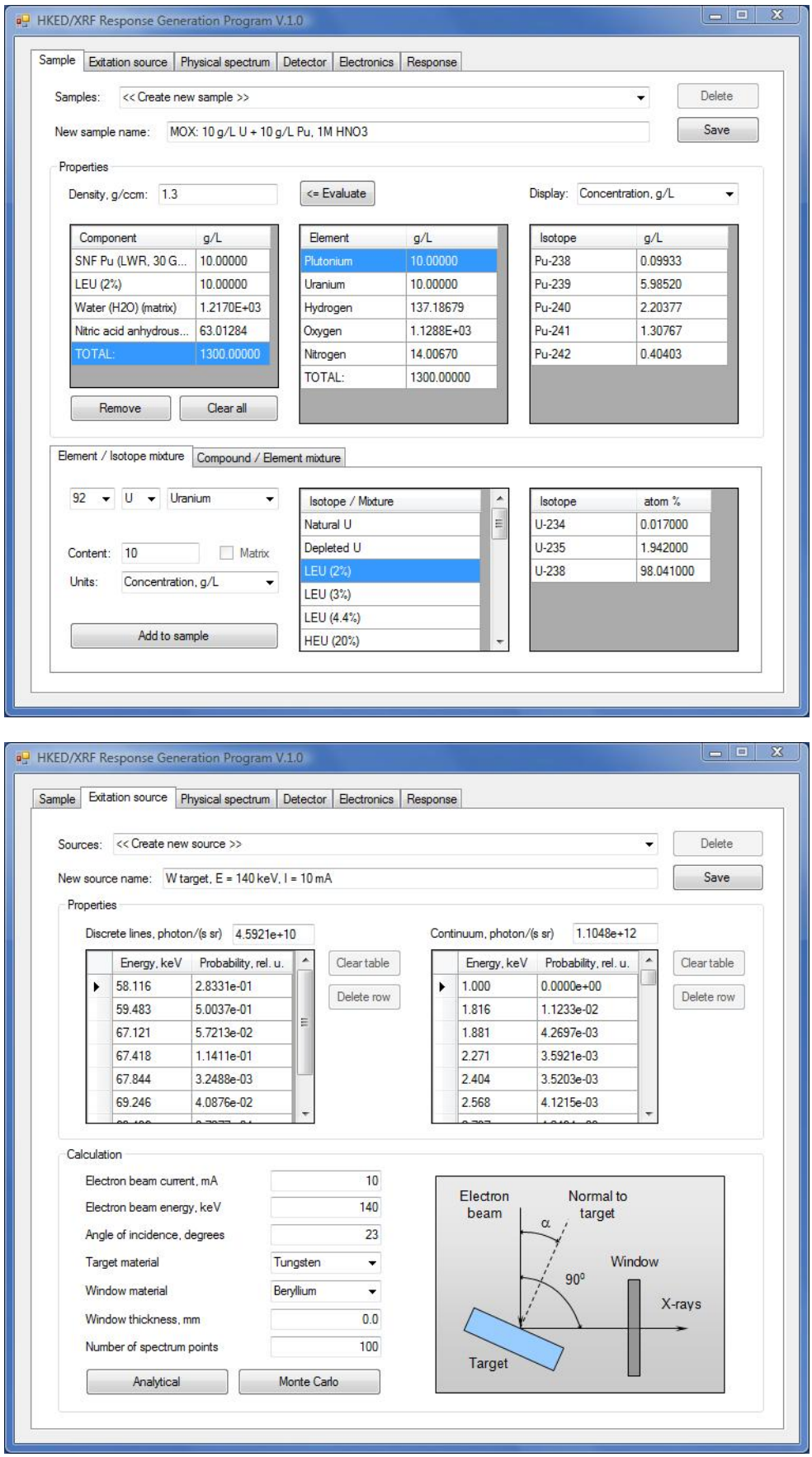

Fig. 2. Screenshots of the HKED-RGP program interface showing the sample editor (on the top) and excitation source modelling (at the bottom) tabs.

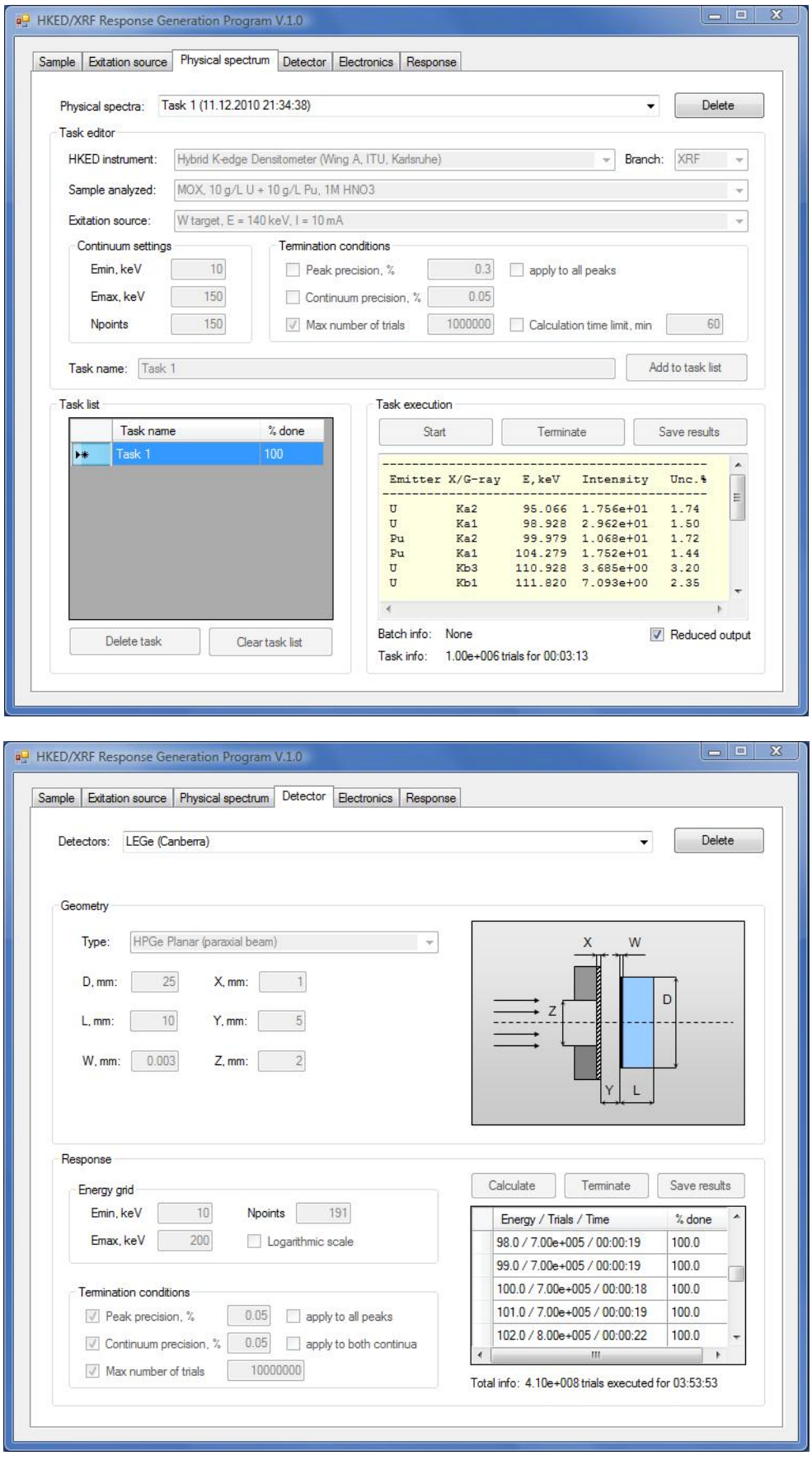

Fig. 3. Screenshots of the HKED-RGP program interface showing the physical spectrum modelling (on the top) and detector characterization (at the bottom) tabs.

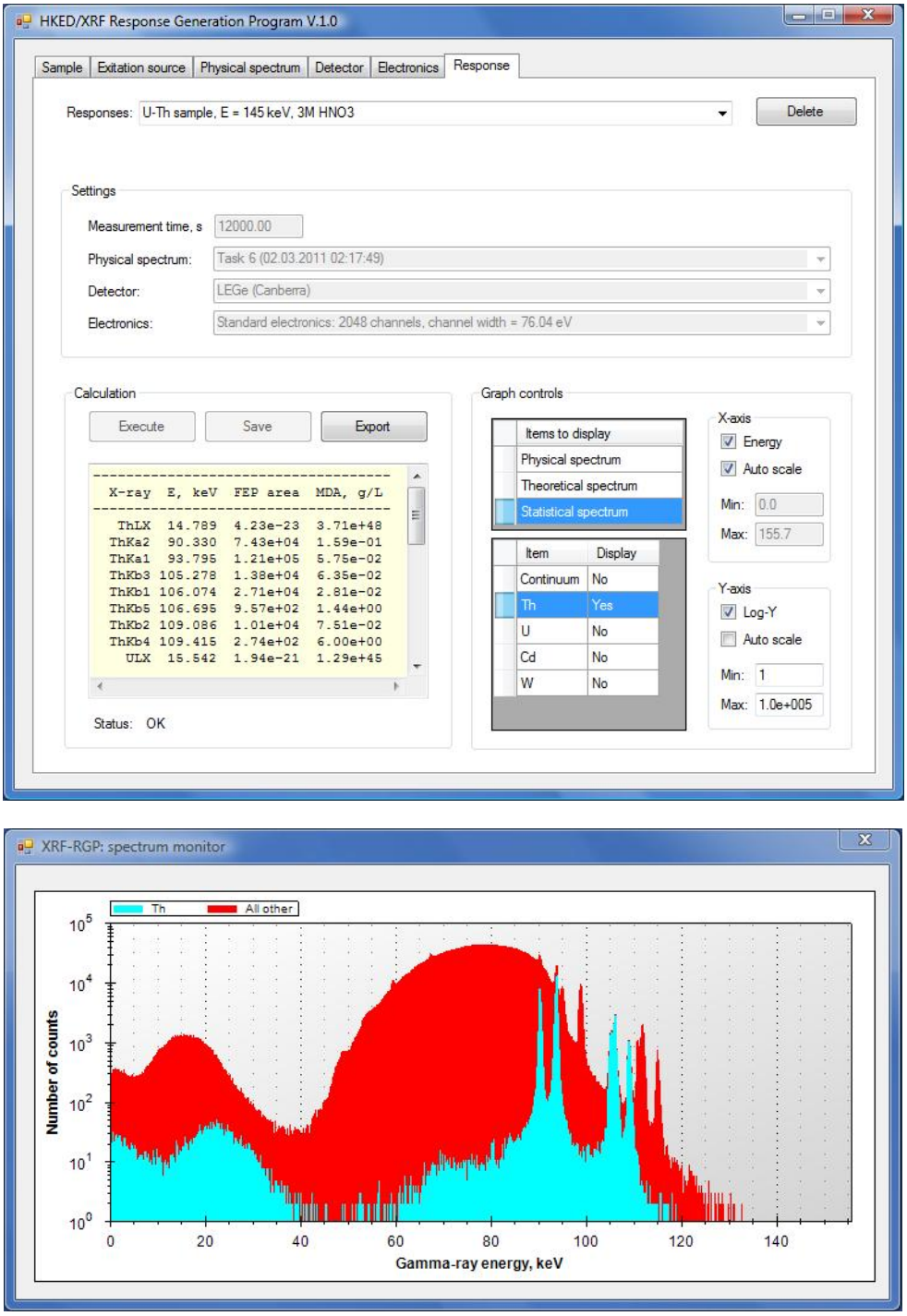

Fig. 4. Screenshots of the HKED-RGP program interface showing the detector spectrum modelling tab (on the top) and a related spectrum monitor window (at the bottom).

2.2. Spectrum modelling approach

The spectrum modelling is performed following a two-step approach described in [3]. In the first step, a physical spectrum at the detector input window is calculated for the specified sample and exciting photon beam using the MCNP Monte Carlo transport engine [4] and an optimized model of a selected HKED system [2]. The spectrum obtained contains a set of discrete energy X/γ-lines and continuum photons resulting from the incoherent scattering of the exciting photon beam and monoenergetic photons in the sample material.

In the second step, firstly, the physical continuum is convoluted with a response matrix, thus forming a major contribution to a continuum part of the resulting spectrum. This is done rather fast as the response matrix generation makes use of the detector responses pre-calculated during the detector characterization. After

that the responses corresponding to the discrete energy X/γ-lines are added. The part of the responses corresponding to the full energy and escape peaks is modelled using the Voigt and Gaussian profiles for X- and γ-lines, respectively. The natural widths of characteristic X-rays in the Voigt profile are estimated based on the widths of the atomic levels involved in the respective intra-atomic transitions. These data are taken from the evaluated atomic data library EADL [5]. The obtained spectrum is finally randomized to mimic the count statistics. No other spectrum distortion effects, e.g. pulse pileup, peak tailing or dead-time losses, are modelled in the present version of the program.

At the output the program generates a comprehensive report in either a text or Excel format that in addition to the modelling results contains also useful analytical data, such as peak areas, background number of counts under peaks, the total and full energy peak efficiencies and the detection limits for each X/γ-peak in the spectrum.

2.3. Upgrade of the X-ray fluorescence database

The accuracy of the XRF spectrum modelling is strongly dependent upon the quality of the photon interaction and X-ray fluorescence data used in the calculation of the physical spectrum. The photo-atomic data used by the MCNP code are provided in the photon transport libraries distributed with the code: MCPLIB, MCPLIB02, MCPLIB03 and MCPLIB04. The use of MCPLIB04 looks beneficial as it contains the most recent evaluation of the photon interaction cross-sections derived from the photon data library EPDL97 [6]. At the same time, regarding the X-ray fluorescence data, it has a number of shortcomings:

- the Kβ-series X-rays are represented by the two effective lines $K\beta'_1$ and $K\beta'_2$ that are the weighted averages of the (M_2-K, M_3-K, M_4-K) and (N_2-K, N_3-K) transitions, respectively. This is a rather rough approximation as with the high-resolution γ-spectrometry the individual Kβ-rays of actinides are normally identifiable in the XRF spectrum;

- the $KO_{2,3}$ and $KP_{2,3}$ X-rays that are easily observed in the fluorescence spectrum of actinides are entirely disregarded;

- the X-ray and K-edge energies as given by MCPLIB04 are shifted to higher values (by about 0.5 keV in the actinide region) compared to the MCPLIB, MCPLIB02, MCPLIB03 and recent X-ray data evaluations [7, 8, 9].

The first two deficiencies result from the X-ray fluorescence sampling scheme defined in [10] and they are common to all versions of MCPLIB. The last deficiency reflects the transition from the conventional fluorescence data of Storm and Israel [11], used in earlier versions of the MCNP photon transport data libraries, to the new EPDL97 based data.

For a check of the inconsistency of the MCPLIB04/EPDL97 X-ray and K-edge energy data, a simple testing experiment was arranged using a 0.12 mm thick lead foil, a ^{109}Cd excitation source and a low-energy planar detector LEGe (Canberra). There were three spectra measured in the following configurations: (i) the ^{109}Cd source shielded with the lead foil, (ii) the same as (i) but without the lead foil, and (iii) the same as (i) but without the source. The test made use of the fact that the single γ-ray of ^{109}Cd (E_γ = 88.0341 keV) lies in a narrow energy interval encompassed by the two PbK-edge energy values as given by MCPLIB03 (E_K = 88.0045 keV) and MCPLIB04 (E_K = 88.290 keV). Therefore, if the MCNPLIB04 data is inconsistent, then the excitation of the PbKX-rays has to take place. The spectra shown in Fig. 5 prove that this is exactly the case.

Based on the indicated findings, a new photon transport library with an improved treatment of the KX-ray fluorescence data was created. For Z > 40 the library distinguishes all 5 fluorescent lines in the Kβ-series. The $KO_{2,3}$ and $KP_{2,3}$ lines were also included for Z > 48 and Z > 80, respectively. The X-ray and K-edge energies were taken from Ref. [7]. All other photo-atomic data, including the cross-sections, excitation probabilities and fluorescence yields, were taken from the EPDL97.

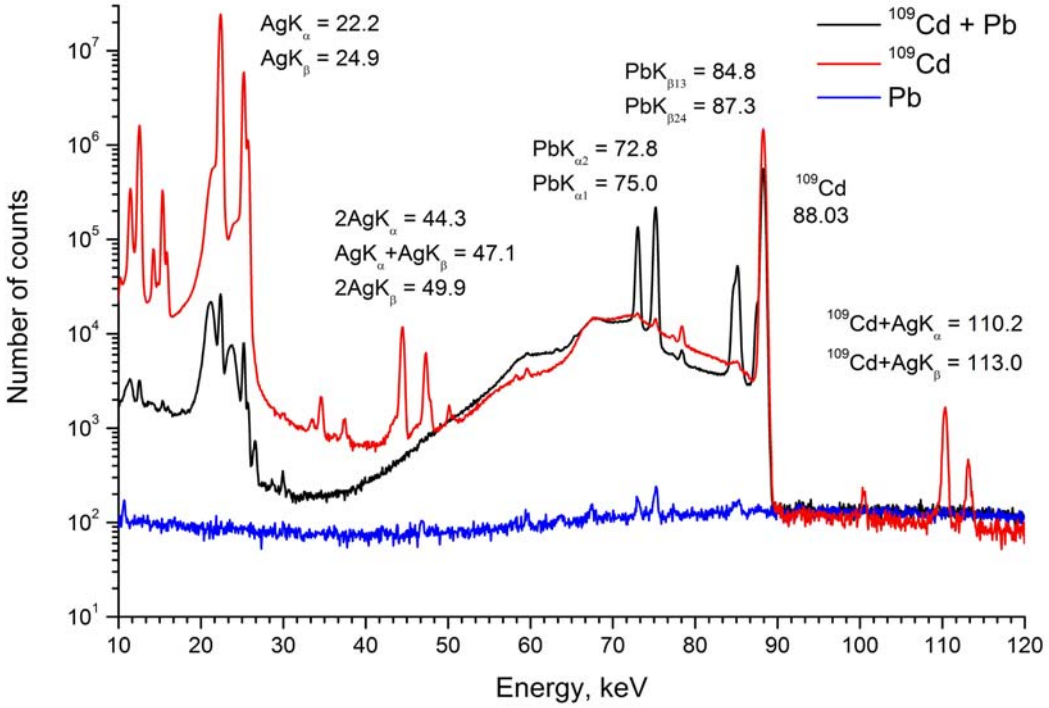

Fig. 5. The results of the testing experiment demonstrating the excitation of the PbKX-rays by the 88.03 keV γ-line of ^{109}Cd.

3. Experimental validation

Within the frames of the current work the experimental validation of the developed software was performed for the XRF channel of an HKED instrument. To this end a number of reference actinide solutions were prepared at the Institute for Transuranium Elements, EC JRC. The measurements were carried out with the HKED instrument available in the ITU's Nuclear Safeguards and Forensics Department.

3.1. Single actinide reference solution

A single low-concentrated actinide solution was intentionally selected for the first validation study as it represents the simplest analytical case allowing easy interpretation and analysis. For the purpose of the study, a reference 2.5M HNO_3 solution containing the 2.026 g/L of Th was carefully prepared. The preparation was done on a gravimetrical basis by the dilution of the Alfa Aesar standard solution with the certified Th concentration at the level of 10016 mgTh/L ± 0.2% (k=2). The density of the obtained reference solution was measured using an Anton Paar densitometer since the knowledge of the experimental density value is essential for Monte-Carlo simulation. This yielded the value of 1.0641 g/mL with the measurement uncertainty ≤ 0.05% (k=2).

To reduce the continuum background under the ThX-ray peaks, the XRF spectrum was collected with a somewhat lower high voltage setting of 145 kV for the X-ray system, which is below the normal setting of 148.8 kV for the X-ray generator as used in routine safeguards measurements. To ensure a sufficient degree of stability for the high-voltage generator (< 0.1%), the X-ray generator was warmed up for three hours prior to the measurement. The actual operating voltage of the X-ray tube was determined to 146.37 kV from the end-point energy E_{max} = 146.37 keV of the excitation spectrum obtained from the K-edge transmission spectrum measured in parallel. This spectrum was also used to monitor the stability of the X-ray beam intensity and of the end-point energy of the bremsstrahlung spectrum throughout the experiment. To provide a sufficient counting statistics in the full energy range, the spectrum was acquired for 3600 s.

The experimental and computed XRF spectra are compared in Fig. 6. Since the absolute intensity of the excitation source was not accurately known, the modelled spectrum was normalized such that the ThKβ_1 peak intensity matched the experimentally observed value. As it is seen from Fig. 6, the simulation agrees reasonably with the experimental data in that it provides a good reproduction of the overall continuum shape

and fluorescent peak intensities. The shortage of counts in the low-intensity spectral regions from 20 to 50 keV and above 100 keV is due to pulse pileup effects that are disregarded in the simulation. Inevitable pulse pileup effects are rather typical for the HKED/XRF measurements that are normally performed at elevated count rates, e.g. 18 kcps for the measurement under consideration.

Another inconsistency between the experimental and simulated spectra is due to the presence of a strong ^{109}Cd source that is built-in in the HKED instrument. The source adds the 88 keV peak and AgKX-ray peaks to the measured XRF spectrum, which are used for the gain stabilization and energy calibration purposes. The source was not included into the simulation as it is not involved in the X-ray fluorescence process of interest and as it further does not interfere with the characteristic X-rays of the actinides.

Fig. 7 provides a closer view (in linear scale) of the Kβ-region of the experimental and modelled fluorescent spectra. It demonstrates the importance of the inclusion of the $KO_{2,3}$ and $KP_{2,3}$ fluorescent lines in the simulation. It also points to a slight disagreement between the theoretical and experimental widths of Th$K\beta_{1,3}$ peaks that are somewhat better resolved in the experimental spectrum. This indicates that still some refinement may be needed in the photo-atomic data used in the simulation, e.g. revision of the intrinsic widths of the $K\beta_{1,3}$ lines.

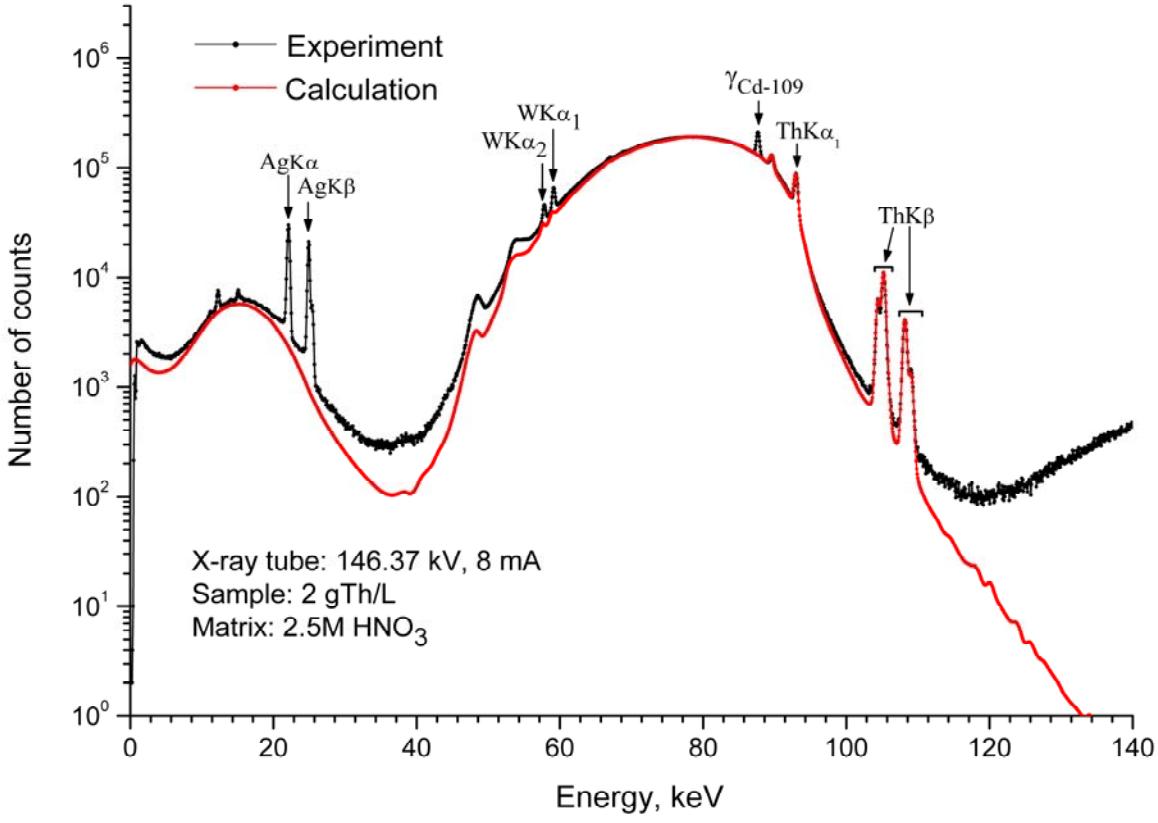

Fig. 6. The experimental and simulated XRF spectra for the 2 g/L Th solution in the 2.5M HNO_3 matrix.

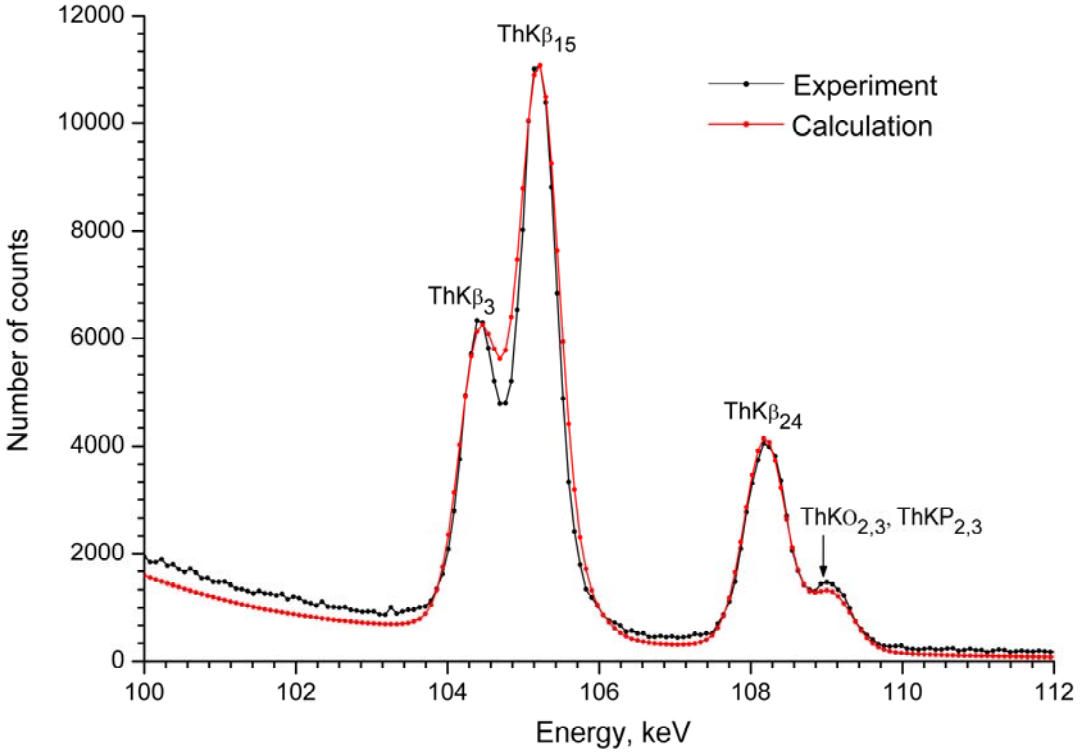

Fig. 7. The Kβ-region of the modelled and experimental fluorescent spectra for the 2 g/L Th solution.

3.2. Safeguards reference U-Pu solution

The routine operations of the HKED instruments in the on-site safeguards laboratories at the reprocessing facilities at La Hague (France) and Sellafield (UK) involve a large number of measurements made on mixed U-Pu solutions. As the support of these measurements is amongst the anticipated practical applications of the modelling software, its validation with this type of samples is rather important.

To this end, a mixed U-Pu 11.75M nitric acid solution was prepared. The U and Pu element concentrations in the solution were determined by isotope dilution mass-spectrometry (IDMS). The IDMS measurements provided the values of 139.319 mg/g ± 0.1% (226.121 g/L ± 0.1%) and 13.382 mg/g ± 0.1% (21.720 g/L ± 0.1%) for the U and Pu element concentrations, respectively. The ^{241}Am content was quantified by high resolution gamma-spectrometry and amounted to 0.127 mg/g (0.206 g/L). The solution density was measured with the Anton Paar densitometer yielding the value of 1.62304 g/mL.

The measurement was performed at the nominal 148.8 kV / 10 mA X-ray generator settings routinely employed for this kind of measurements. The actual X-ray tube acceleration voltage was determined experimentally to 150kV from the end-point energy of the bremsstrahlung continuum in the transmission spectrum accumulated in parallel. The spectrum was acquired for 2000 s live time to ensure good statistics of counts in the full energy range.

The experimental and modelled spectra are compared in Fig. 8. For the sake of the comparison the modelled spectrum was normalized such that the intensity of the UKβ$_1$ peak matched the peak intensity in the measured spectrum. The agreement between the simulation and experiment is satisfactory. The origin of the observed deviations between the two spectra is essentially the same as already discussed in section 3.1, i.e. the pulse pileup effects and the contribution from the ^{109}Cd calibration source.

Another disagreement between the experimental and simulated XRF spectrum can be noted for the intensity of the WKX-ray peaks, which are somewhat underestimated in the simulated spectrum. The tungsten X-rays are mainly created in fluorescence events occurring in the tungsten collimator placed between detector and sample vial. Not very much attention has been paid in the modelling to this kind of “parasitic” fluorescence effects in construction materials of the instruments because they are of no practical relevance for the prime

purpose of the model, i.e. obtaining an accurate fluorescence response from the sample analysed rather than from the instrument's construction materials (see [2]).

Fig. 9 zooms in the Kβ region of the simulated and experimental spectra, demonstrating an almost ideal agreement between the two in this complex region. A possible origin of a minor deficiency observed in the $UK\beta_{13}$ doublet was already discussed in section 3.1.

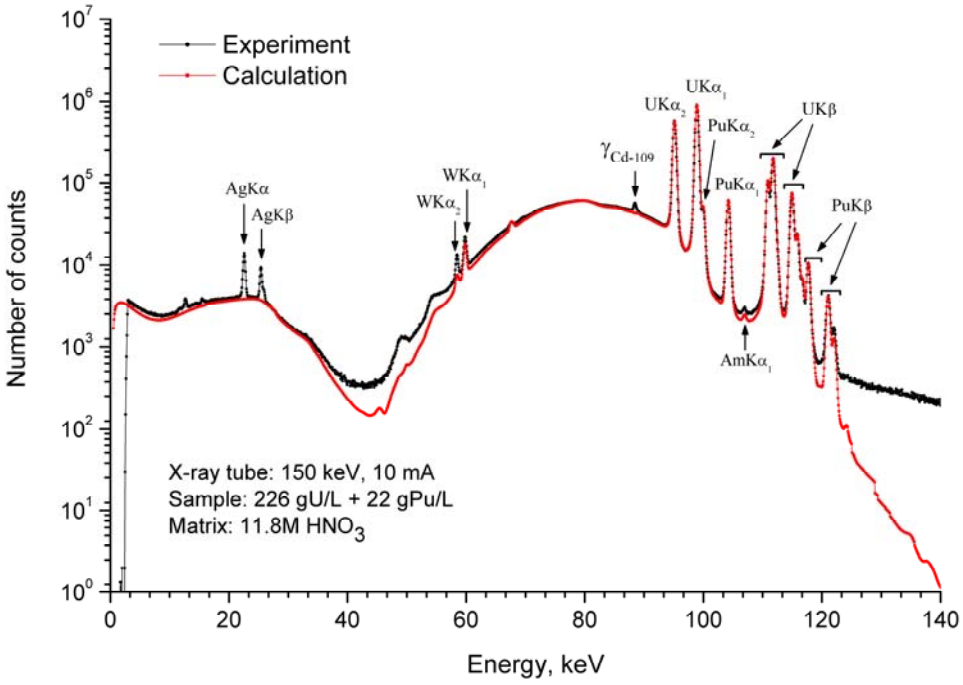

Fig. 8. The experimental and simulated XRF spectra for the mixed U-Pu solution containing 226 gU/L and 22 gPu/L in 11.8M HNO_3 matrix.

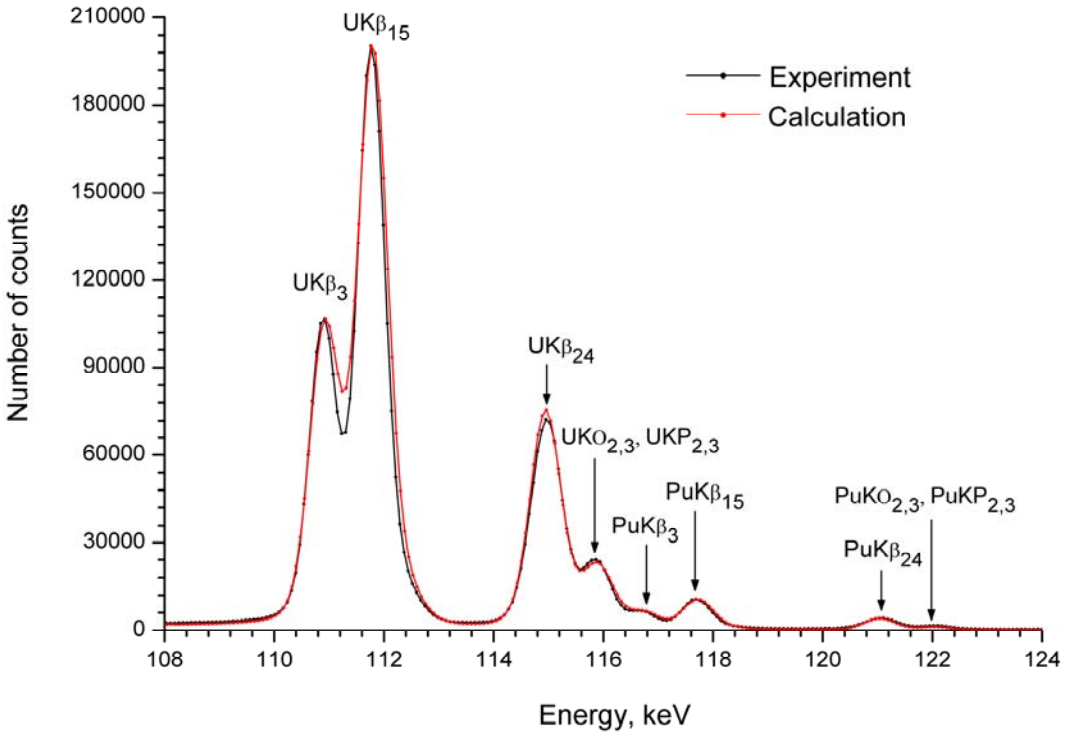

Fig. 9. The Kβ region of the measured and modelled XRF spectra from the mixed U-Pu solution.

3.3. Reference solutions of non-safeguards origin

The necessity of analyzing different actinide mixtures arises rather often in the nuclear fuel cycle research activities carried out in ITU. These samples may contain actinides in a wide concentration range and in various matrices that differ significantly from the typical safeguards samples in reprocessing plants. For this kind of special samples the use of the modelling approach is almost inevitable to avoid huge calibration efforts otherwise needed for reasonable instrument calibration. This is another important area where an extensive application of the developed modelling software is expected.

In the first study, a rather unusual actinide mixture containing approximately equal concentrations of U and Th was considered. A respective reference solution was prepared on a gravimetrical basis from the certified Alfa Aesar Th standard (see section 3.1 for details) and a reference U solution. The latter was obtained by dissolving the IRMM EC110 UO_2 sintered pellet in the 3M nitric acid. The resulting concentrations of U and Th in the prepared solution were 2.4349 g/mL and 2.4077 g/mL, respectively. The resulting molarity was 2.6. The solution density was determined to 1.08771 g/mL using an Anton Paar densitometer. The experimental spectrum was collected at X-ray generator settings of 145 kV / 8 mA (again, the actual high voltage was 146.37 kV as before) for a live time of 3600 s. The experimental and modelled spectra are compared in Figs.10 and 11. The normalization of the simulated spectrum was done such that the intensity of the Th$K\beta_1$ peak matched the experimentally observed value.

In the second study, reference samples containing low actinides in a heavy matrix were examined. Such samples originate from the on-going experiments on the pyrochemical spent fuel processing technology development. The matrices in these samples are usually represented by molten salts (LiCl, KCl) or heavy metals (Zr, Cd, Bi) that are completely different from the matrices encountered in the conventional PUREX process.

For the purpose of the study, two 4M HNO_3 reference solutions with the same concentration of U (1.14 g/L) and Pu (1.17 g/L) were prepared. The preparation was done gravimetrically by dissolving pure ingots of Cd (sample 1) and Bi (sample 2) in a certified stock-mother U-Pu solution. The resulting concentrations of heavy metals were 110.5 gCd/L and 117.2 gBi/L. The solution densities were determined using the Anton Paar densitometer, yielding values of 1.26203 g/mL and 1.2722 g/mL for the Cd-matrix and Bi-matrix solutions, respectively. The spectra were acquired for the 5000 s live time presets at the 148.8 kV / 8 mA X-ray generator settings. The actual X-ray tube acceleration voltage was determined experimentally to 150 kV from the end-point energy of the bremsstrahlung continuum in the transmission spectrum accumulated in parallel. The obtained experimental and modelled spectra are compared in Figs. 12 and 13. The modelled spectra were normalized such that the intensities of the Pu$K\alpha_1$ line match the corresponding intensities in the experimental spectra for the Cd- and Bi-matrix samples.

Apart from the influence of the pulse pileup effect which could be explained by high counting rates of 31kcps for Bi-matrix sample and of 15kcps for Cd-matrix sample that severely affects the agreement in the low-intensity regions of the spectra, the most important features of the fluoresced spectra are perfectly reproduced. These include shapes of the continuum bumps that suffer a drastic change when switching from the light to heavy matrix solutions as well as the fluoresced peak intensities of both actinides and matrix elements.

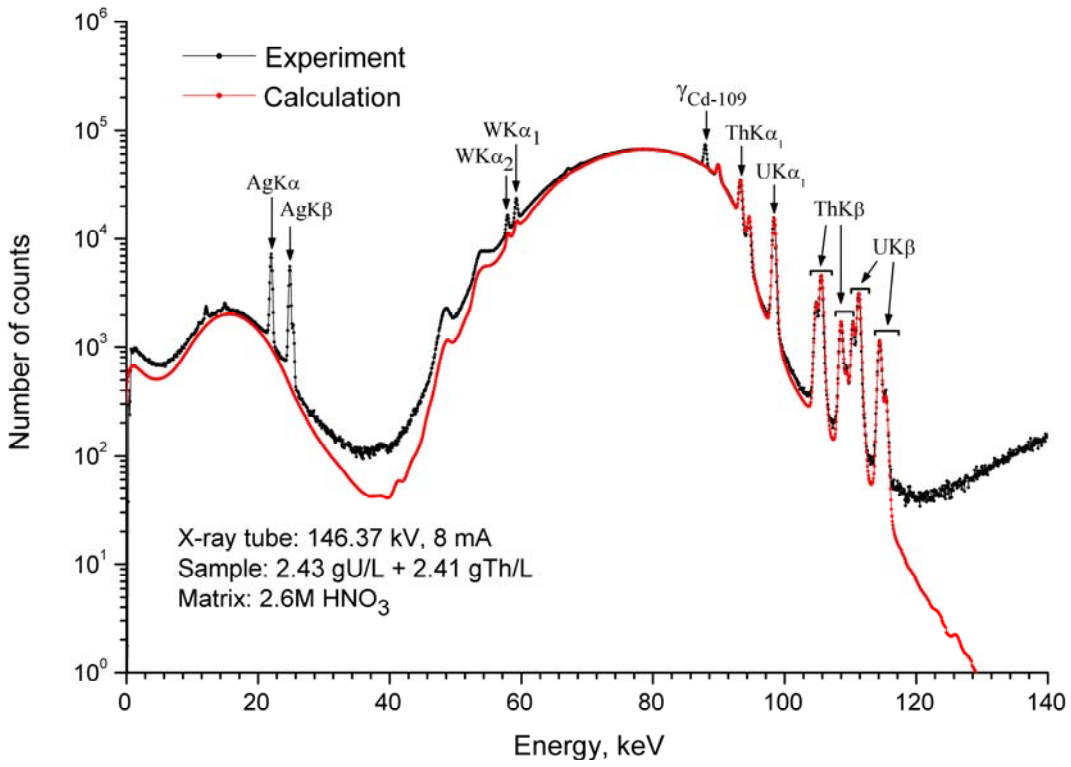

Fig.10. The experimental and simulated spectra for the low-concentrated U-Th solution containing 2.43 gU/L and 2.41 gTh/L in the 2.6M HNO_3 matrix.

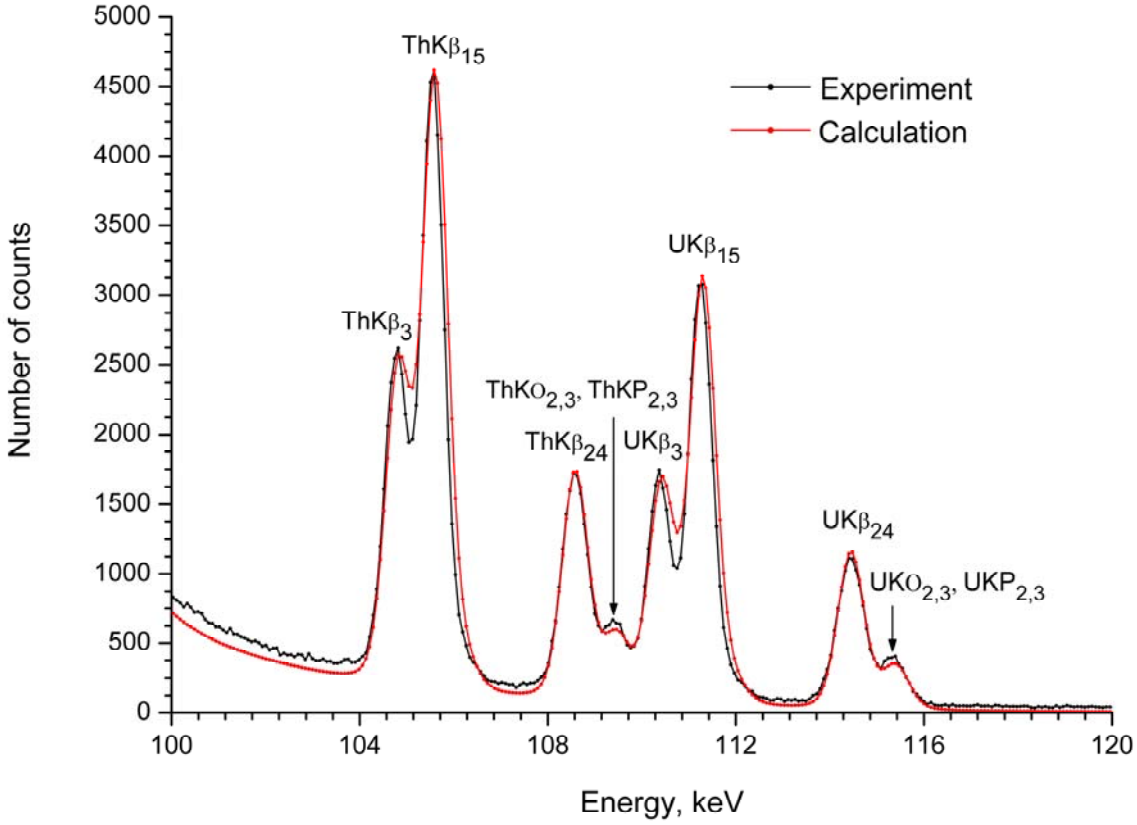

Fig. 11. The Kβ-region of the experimental and simulated spectra for the U-Th reference solution.

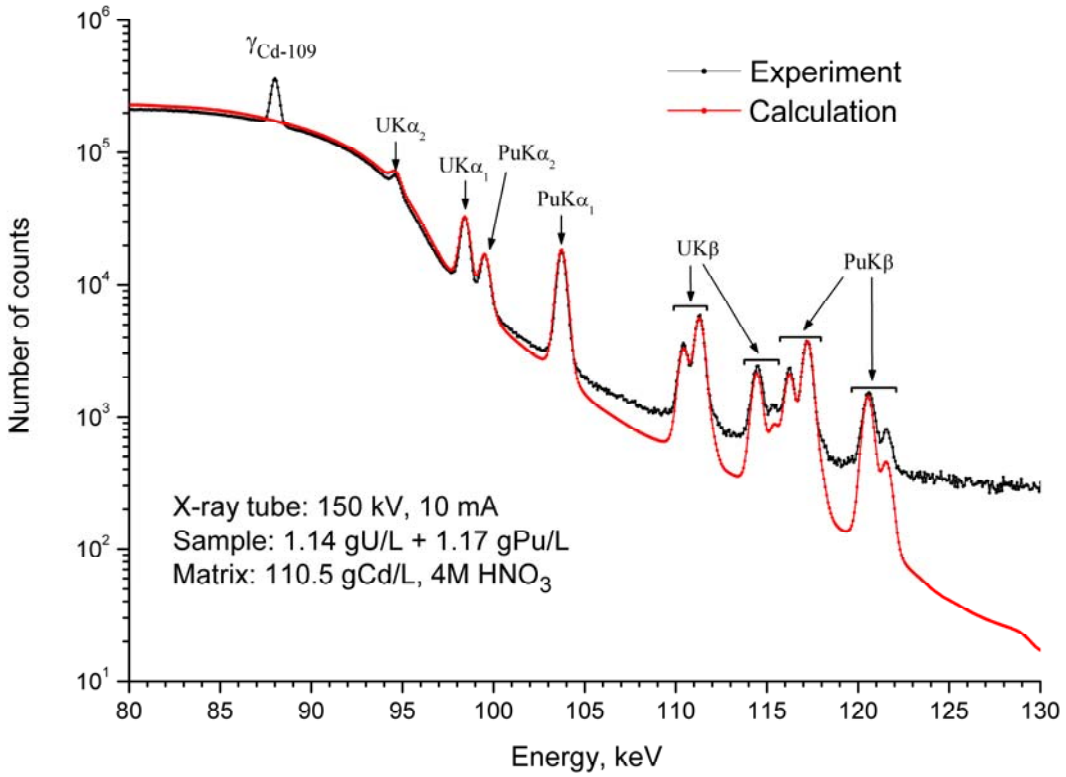

Fig. 12. The experimental and simulated spectra in the energy range of 80-130 keV for the low-concentrated U-Pu solution containing 1.14 gU/L and 1.17 gPu/L in the 110.5 gCd/L 4M HNO_3 matrix.

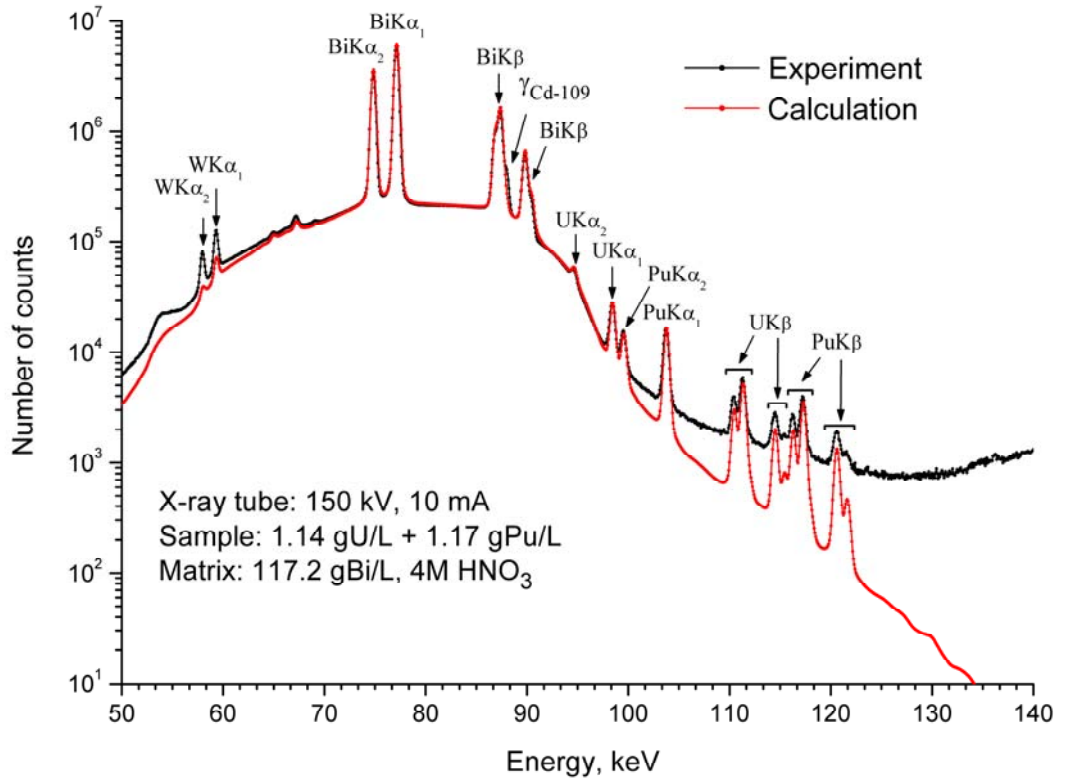

Fig.13. The experimental and simulated spectra in the energy region of 50-140 keV for the low-concentrated U-Pu solution containing 1.14 gU/L and 1.17 gPu/L in the 117.2 gBi/L 4M HNO_3 matrix.

4. Summary and conclusions

The HKED-RGP simulation program developed offers an integrated solution for modelling all aspects of the HKED measurements. The program is capable of predicting the response of hybrid K-Edge/K-XRF instruments in terms of the X/γ-ray peak intensities and full spectrum. It is purely based on the fundamental physics and does not require a special calibration or any external data other than the instrument geometry, the X-ray generator operational settings, the elemental composition and density of a sample analyzed and detector parameters. The normalization of the absolute X/γ-ray peak intensities and full spectrum is however required to take account of the actual intensity of the excitation source. All measurement branches including the transmission, XRF and passive gamma channels can be modelled using the same user-friendly simulation environment offered by the program.

The results of the on-going validation effort presented in this paper demonstrate a reasonable agreement between the experimental and simulated spectra regarding their most significant features, such as the fluoresced peak intensities and the continuum distribution shapes. The accuracy of the photo-atomic data was shown to play a crucial role for the quality of the simulation. A new photo-atomic transport data library was created to allow a more correct treatment of the X-ray fluorescence process as modelled by Monte Carlo. As a result of the validation a few minor inaccuracies in the simulation were identified, e.g. due to the disregard of the pulse pileup effect and inaccuracies in the atomic relaxation data. These will be addressed in the future work.

The HKED-RGP is aimed to serve as a mathematical calibration tool for the HKED measurements. This particularly implies that after the completion of the comprehensive experimental validation, the modelling approach will be used in the quantitative interpretation of the measured spectra, thereby drastically reducing the calibration effort. It may also be very useful in testing the HKED spectrum processing algorithms, especially for non-common actinide mixtures and complex matrices, for which the appropriate standard materials are not always readily available in analytical laboratories. It is intended ultimately that the HKED-RGP will serve as a simulation utensil in the Monte Carlo aided XRF spectrum processing, e.g. using the Monte Carlo least squares method described in [12]. This approach will be especially useful for the accurate background delineation under the full energy peaks in the XRF spectrum that represents a major problem when achieving a relative accuracy better than 0.5% is required. After experimental validation, the modelling approach will be used to interpret experimental spectra measured, for example, under variable matrix conditions. This will drastically reduce calibration efforts.

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LB-NA-24-882-EN-N

DOI: 10.2760/07768