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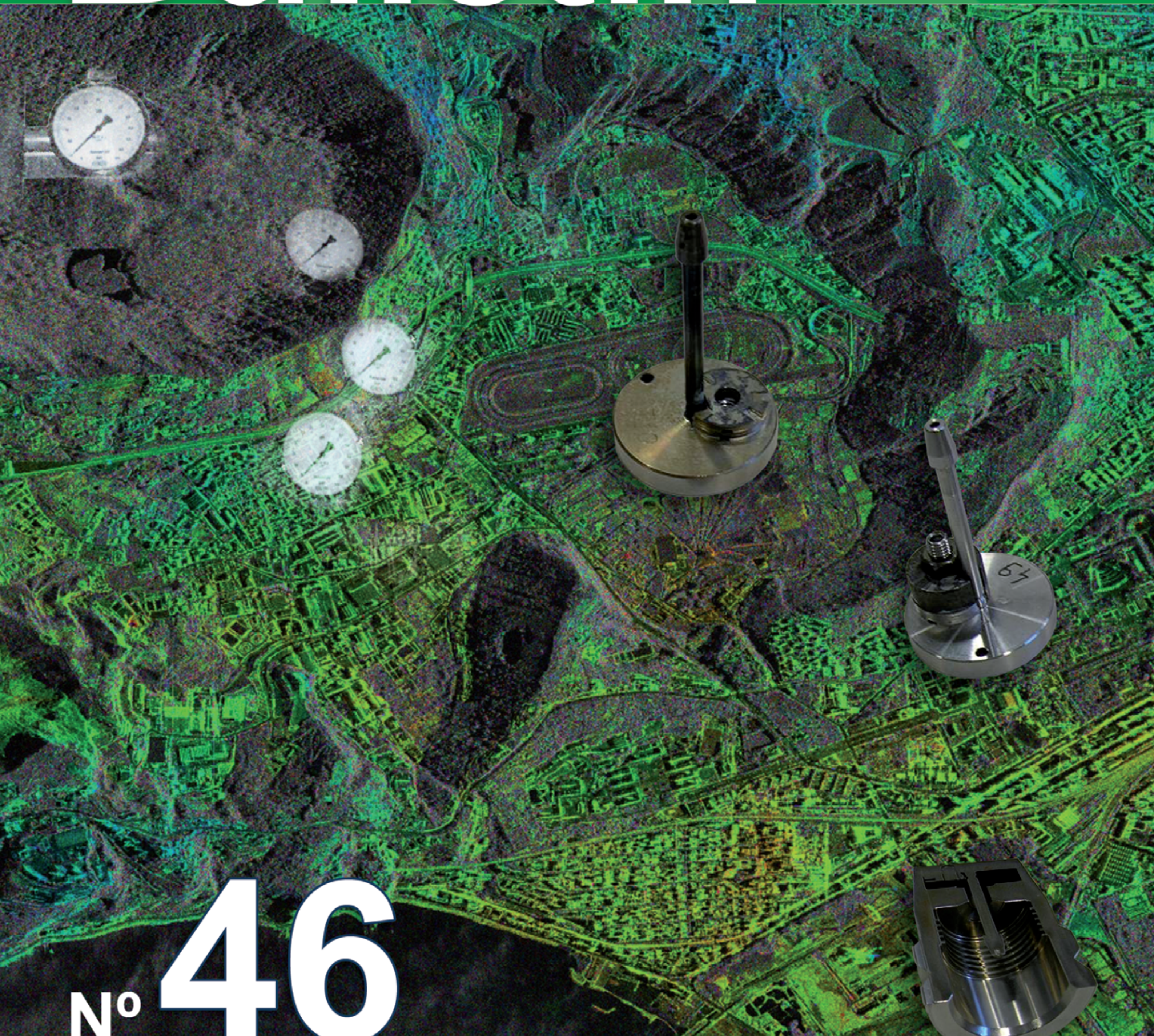
Bulletin



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ESARDA
European Safeguards Research and Development Association

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ESARDA News

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 yEuropean 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 International Atomic Energy Agency (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.

The EC-SP is an integral part the European Union's nuclear non proliferation policy [1]. Within the framework of the Euratom Treaty (1957), the European Commission's Directorate General for Energy (ENER) implements a European Union wide Regional System of Nuclear Material Accountancy and Control (RSAC). The Joint Research Centre – JRC, a sister Directorate General from the European Commission, provides, among others, the research, development and technical support to this RSAC and to the IAEA. JRC technical activities contribute to the improvement of the implementation of Nuclear Safeguards and, in a wider view, to the implementation of nuclear non-proliferation policies.

Beyond JRC and ENER, other European Commission services get inspiration from the EU non proliferation policy and regularly provide funding to IAEA specific projects. A good example is the EU support to the IAEA ECAS project – "Enhancing the Capabilities of IAEA Analytical Services" – from the Instrument for Stability. In such cases, the JRC, via the EC-SP, can provide the necessary scientific/technical assistance to the relevant Commission Services closing the gap between financing authority and the end-user.

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 DG 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 scientific and technical 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 Materials and Measurements (IRMM), Geel, Belgium
- Institute for Transuranium Elements (ITU), Karlsruhe (Germany) and Ispra (Italy) sites

The European Commission Directorate General for 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.

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, often together with colleagues from ENER, regularly participate 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. Analysis of Nuclear Materials and Environmental Particle Samples – NWAL

The support to IAEA also includes the analysis of nuclear materials, of environmental particle samples, and the provision of reference/QC materials. These activities are performed in JRC laboratories in the frame of IAEA's Network of Analytical Laboratories (NWAL).

3.4. Scientific and Technical Support to EC Services supporting the IAEA

When other European Commission services support the IAEA, as part of the European Union non-proliferation policy, the EC-SP can be called to provide the necessary scientific and technical assistance to the relevant Services closing the gap between financing authority and the end-user.

3.5. 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 it is also possible to better coordinate and focus on future efforts and initiatives.

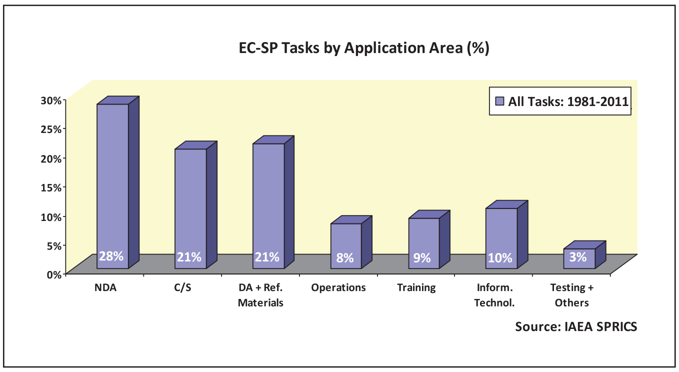

Figure 1: Distribution of EC-SP tasks along the different Safeguards technical and application areas for the period 1981-2011.

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 as well as recognised observers. Within this context, 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 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.

In Autumn 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 (e.g., 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

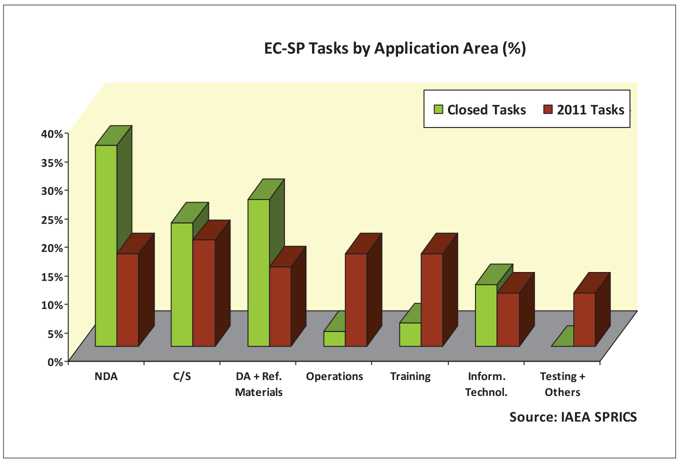

Figure 2: Distribution of EC-SP closed and active tasks in terms of the Safeguards technical and application areas

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.

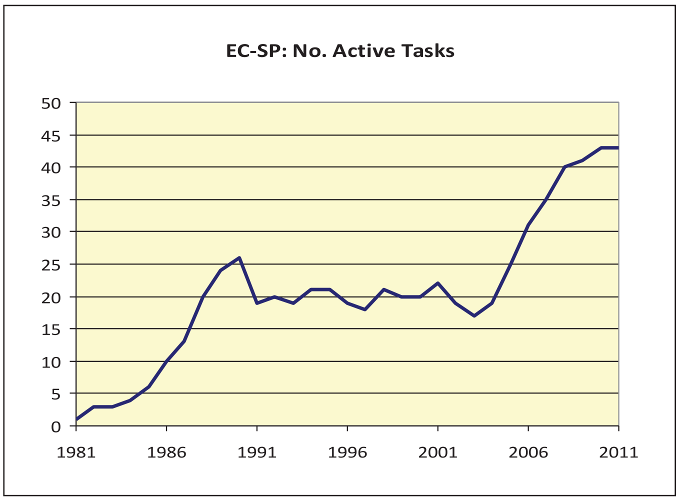

Figure 3: Number of EC-SP active tasks.

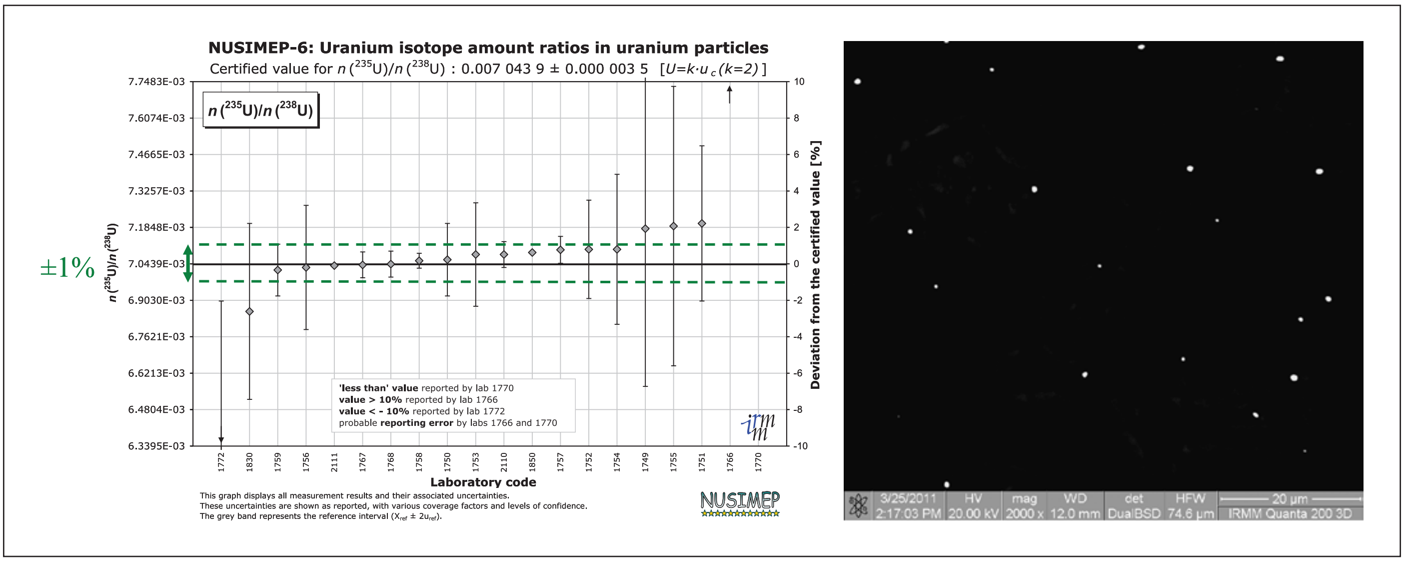

Figure 4: NUSIMEP-6 participants results on isotope amount ratios in uranium reference particles

5.1. Reference Materials

EC A 00318 – Special Reference and Source Materials for Destructive Analysis has been one of the first EC support tasks to the IAEA and is still ongoing today. Particularly appreciated by the IAEA were the recently produced series of IRMM uranium isotopic reference materials containing ^{236}U. Amongst those are the IRMM-3636 $^{233}U/^{236}U$ double spike and the IRMM-3100a $^{233}U/^{235}U/^{236}U/^{238}U$=1/1/1/1 "quad" isotopic reference material, suitable to perform internal mass fractionation corrections and verifying the inter-calibration of multi-detector systems in isotope mass spectrometry. As a result IRMM certified reference materials are now regularly applied by IAEA's new Office for Safeguards Analytical Services – SGAS, strengthening the effectiveness and efficiency of IAEA Analytical Services.

Another EC support task from the early beginnings of the EC SP to date is EC A 00267 – Analytical Quality Control Services. NUSIMEP is an external quality control programme organised by IRMM with the object of providing materials for measurements of trace amounts of nuclear materials in environmental matrices. The most recent IRMM Interlaboratory Comparisons (ILCs) under this support task are NUSIMEP-6 and NUSIMEP-7 organised for DG ENER and the IAEA Network of Analytical laboratories (NWAL) on uranium isotope amount ratios in uranium particles [2]. Participation in NUSIMEP enables the NWALs to demonstrate their measurement capabilities on uranium reference particles, similar to the ones found by inspectors in environmental swipe samples, to safeguards authorities.

Recently, under the initiative and coordination of IRMM, four key nuclear mass spectrometry laboratories (IRMM, ITU, IAEA-SGAS and 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 [3]. MTE provides a measurement performance which is superior compared to the present IAEA requirements, enabling more detailed conclusions from measured sample data for source attribution. The performance of the MTE method for the minor uranium ratios n(^{234}U)/n(^{238}U) and n(^{236}U)/n(^{238}U) can be seen in Figure 5. The capabilities and sample throughput for measurements of the minor uranium isotope ratios have been improved by implementing MTE at the IAEA SGAS-Seibersdorf under the task EC-B-01752 on advanced training techniques in Mass Spectroscopy. Due to the excellent performance of this technique MTE is now officially accepted for safeguards measurements at the IAEA SGAS.

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-01806 – Verification of mixed U-Pu Spikes] [4]. The reference materials used to accomplish

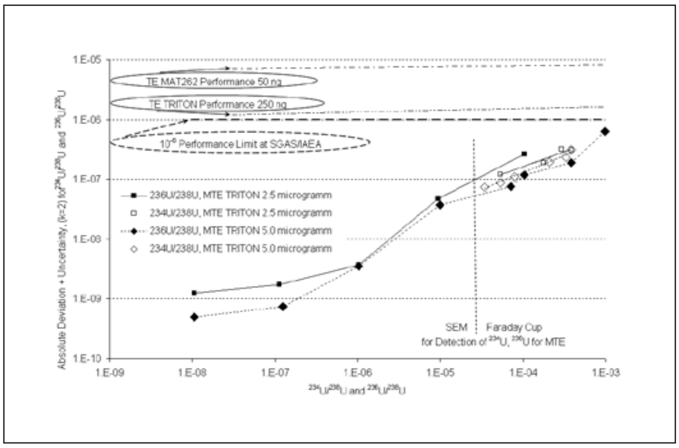

Figure 5: Performance of the Modified Total Evaporation method for minor uranium ratios n(^{234}U)/n(^{238}U) and n(^{236}U)/n(^{238}U)

this task were subject to an inter-calibration campaign to underpin the confidence in the use of IRMM plutonium spike 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 to the SI was confirmed [5].

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 are 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 [6]. 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.

JRC-ITU is currently in the process of acquiring a Large Geometry Secondary Ion Mass Spectroscopy instrument – LG-SIMS (with co-financing from DG-Energy). This instrument is the very same as the one that the IAEA has installed 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

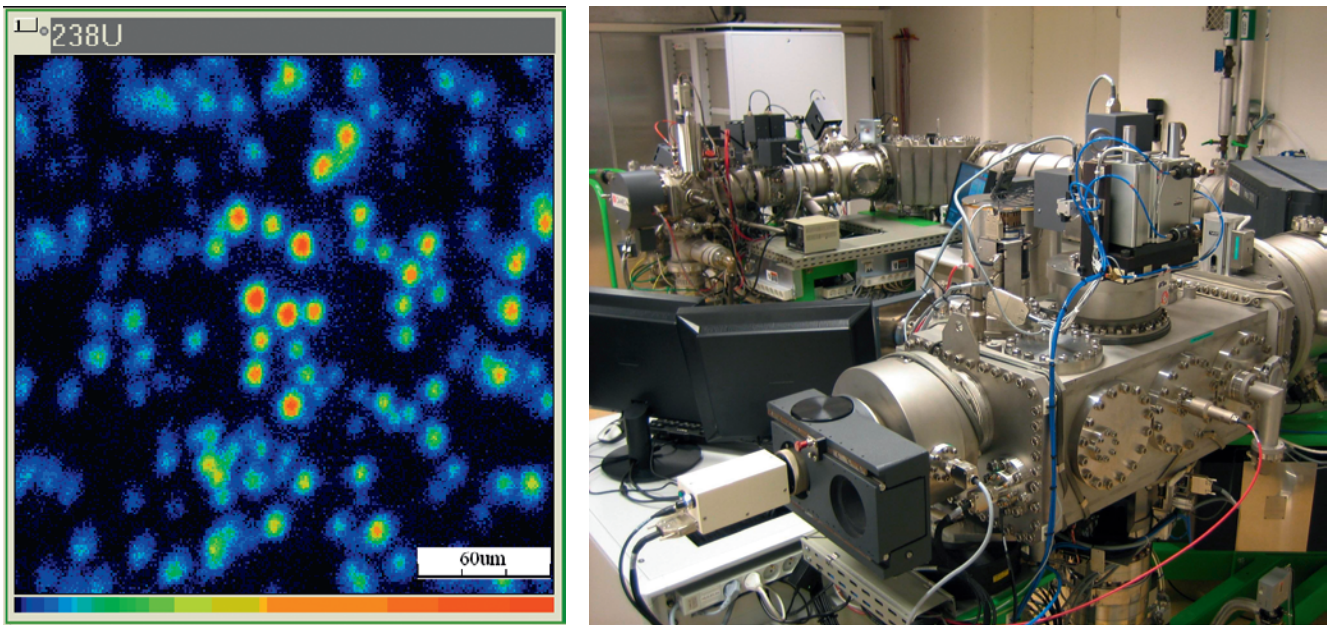

Figure 6: Example of JRC-ITU Automated Particle Measurement (APM) screening software that was recently developed in cooperation with the company Cameca , and a photo of an LG-SIMS from NORDSIM laboratory, Stockholm (equal to the one to be soon installed at JRC-ITU).

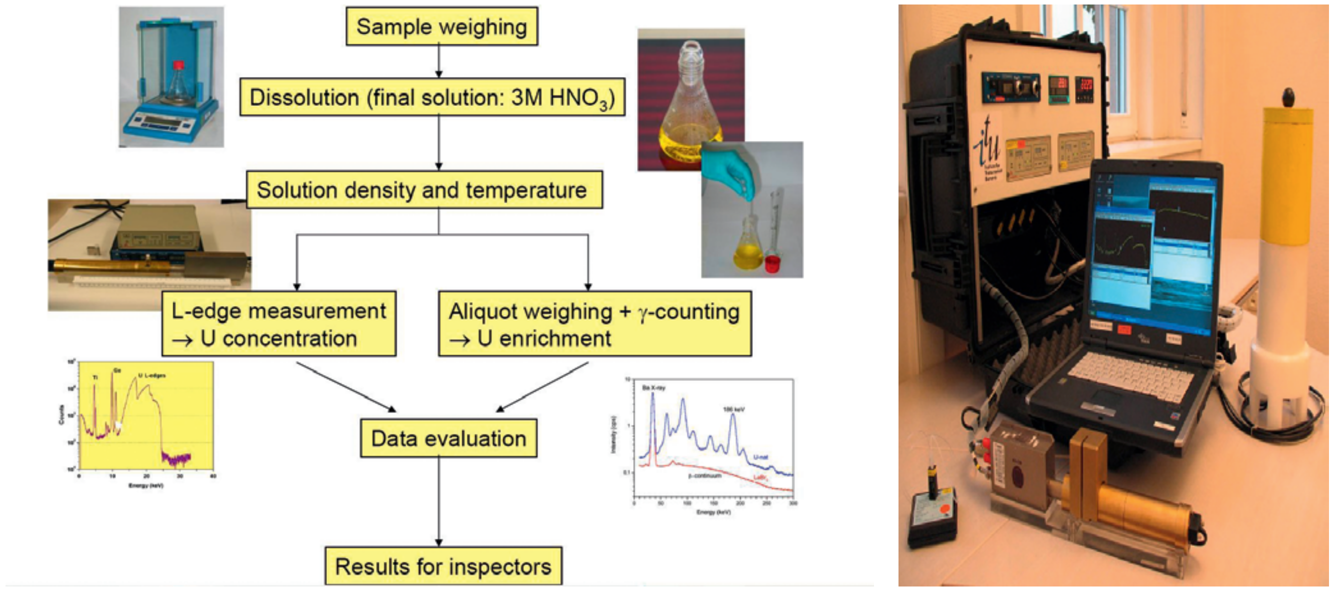

Figure 7: Procedure for the COMPUCEA technique and equipment

NaIGEM code is used, which has recently been adapted to handle the presence of reprocessed uranium.

Following the successive and extensive evaluation of COMPUCEA's performance [7, 8, 9], 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 [10]. 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

Figure 8: Core of ultrasonic seals.

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.

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 [11]. The DIV system [12] 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.

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, Euratom, states 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 pa-

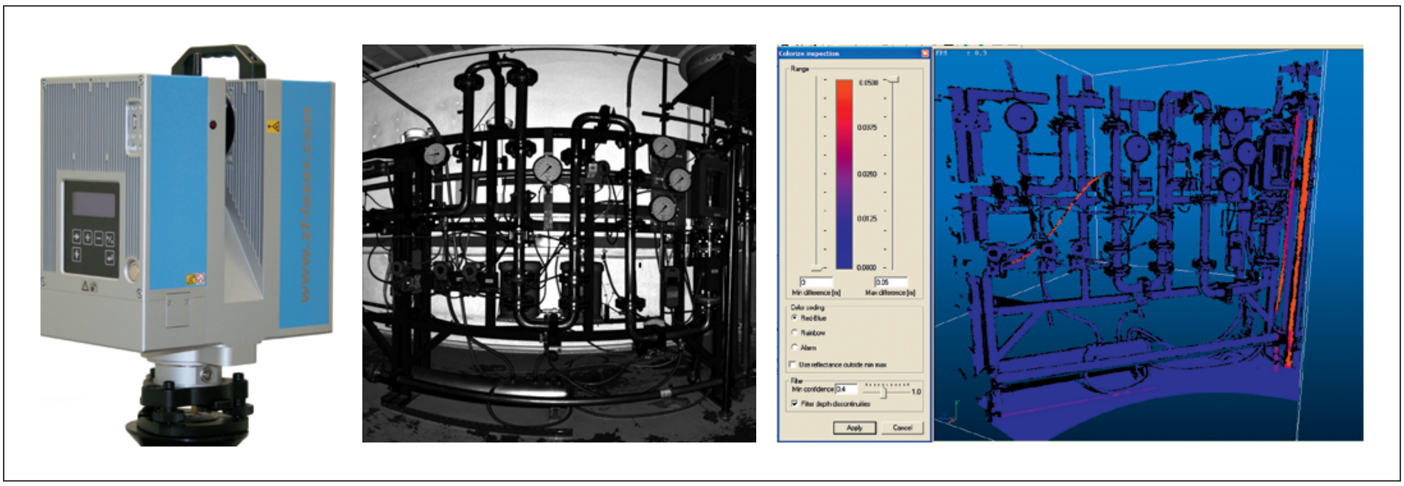

Figure 9: 3D Laser Range Finder used in the 3DLR, and examples of 3D capturing of a complex scene including automated 3D scene change detection – for verification purposes.

Figure 10: L2IS Portable and Fixed reading Stations

rameters 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 [13].

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 [14]. 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.

5.7. Support to IAEA ECAS project

The European Union (EU) has affirmed that it will support international cooperation on technological infrastructure and networks necessary to verify the non-diversion of declared nuclear material but also the absence of illicit nuclear material and activities. The EU envisages supporting the ongoing efforts to strengthen IAEA's analytical capabilities with a contribution from the Instrument for Stability (IfS) to the expansion and modernisation of the IAEA Safeguards Analytical Laboratories (SAL) under the project of "En-

Figure 11: H. Nackaerts IAEA Deputy Director General, Head of the Department of Safeguards, Y. Amano IAEA Director General, Y. Aregbe EC-JRC-IRMM and G. Voigt Director IAEA-SGAS at the groundbreaking ceremony for the new IAEA Nuclear Material Laboratory, Seibersdorf, 7 Sept. 2011.

hancing Capabilities of the Safeguards Analytical Services" (ECAS). On requests of EuropeAid Development and Cooperation (DG DEVCO) and the European External Action Service (EEAS) the JRC provides via the EC-SP technical/scientific advice for the EU donation for the new IAEA Nuclear Material Laboratory. The groundbreaking ceremony (Figure 11) was held on 7 September 2011 at the International Atomic Energy Agency -Safeguards Analytical Laboratories –IAEA-SGAS, Seibersdorf.

6. Discussion and Conclusions

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

In recent years, EC-SP contributions have expanded from activities in research and development in Nuclear Safeguards basic disciplines – C/S, DA and NDA, and now also include 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 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" in both Safeguards and Non-Proliferation issues.

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 authors express their 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 the important role of execution of the different tasks, as well as for having provided material for this paper.

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The authors also acknowledge 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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URENCO, an international success story

URENCO is an independent, international energy and technology group operating in the enrichment stage of the nuclear fuel supply chain. With production from facilities in Germany, the Netherlands, the UK and the US, URENCO's focus is on providing safe and reliable uranium enrichment services for civil power generation, within a framework of high operational and environmental standards.

URENCO is an international success story. The Treaty of Almelo, signed by the governments of Germany, the Netherlands and the UK on 4 March 1970, established essential principles for the effective supervision of URENCO's technology, centrifuge manufacturing and operations. Since the signing of the Treaty, URENCO has fully justified the negotiations and decisions of 1970. Four decades on, the agreement remains the cornerstone of international collaboration on non-proliferation, while URENCO has grown considerably and extended its international relationships to include the US and France. URENCO's foundations provide a model of how international co-operation can ensure a safe, secure and commercially attractive supply of nuclear fuel for the peaceful production of nuclear power.

URENCO is a leading supplier in the global enrichment market, with demand for the Group's services continuing to increase. Supported by strong demand, during 2010 the Group achieved a further increase in the forward order book to €21 billion, extending beyond 2025.

A major achievement in URENCO's history came in June 2010 with the inauguration and start of commercial operations at our new enrichment plant in the USA. This milestone event was the culmination of several years of extensive infrastructure and plant investment, and represents a significant accomplishment for both the organisation and the wider nuclear industry.

URENCO is committed to sustainable operations and endeavours to be a good corporate citizen. Decision-making across all business areas prioritises corporate responsibility, and the Global Reporting Initiative standards are used to benchmark our performance in this area.

Integrity, one of URENCO's company values, attaches a great importance to a dedicated Safeguards culture within the company. Enrichment is one of the most proliferation-sensitive parts of the nuclear fuel cycle which needs an effective non-discriminatory Safeguards regime. Non-proliferation aspects are considered throughout all company business areas, from contract negotiations through to implementation of operational procedures.

Safeguards by Design has always been considered beneficial by URENCO for all stakeholders including operators of nuclear facilities if applied with the necessary sense of proportion. It has been practised by close communication with the International Safeguards organisations and by applying the Safeguards experience gained over many years.

URENCO has decades of experience of the development and implementation of Safeguards regimes in Gas Centrifuge Enrichment Plants (GCEPs). The current safeguards regime for GCEPs, which is based on the Hexapartite Safeguards Project and the Additional Protocol and recently improved by the Partnership Approach between Euratom and the IAEA, was substantially shaped by active URENCO contributions.

URENCO has a strong interest in sharing Safeguards experience with other stakeholders. This is regularly done by participation in and active contribution to international Safeguards conferences as ESARDA, INMM and GCEP-specific international workshops.

In 2009, URENCO organised a GCEP-specific Safeguards conference in Chester, UK which has been attended by 77 people from 13 countries worldwide. The aim of the conference was to discuss and influence the Safeguards strategies in GCEPs. A wide range of worldwide Safeguards experts from GCEPs, Governments, Safeguards Authorities, Research Organisations and Universities discussed in working groups how to detect undeclared materials and activities as well as the right balance between remote monitoring and inspector presence.

GCEP-specific training of inspectors is essential for effective Safeguards inspections in GCEPs. URENCO supports EURATOM and the IAEA with regard to GCEP specific inspection training with particular focus on LFUAs[1]. This support is aimed at optimising the inspection process, inter alia by taking operator aspects into account.

New Safeguards strategies are often accompanied by the application of new techniques. Before such techniques come into regular operation, they have to be tested by taking all aspects into account including operator requirements. URENCO has hosted many field trials for new techniques over the years. Many of these field trials have led to routine safeguards application.

URENCO looks forward to continue contributing to efficient, effective and non-discriminatory Safeguards systems, and in 2011 is now officially a member of ESARDA.

[1] LFUA= Limited Frequency Unannounced Access involves restricted inspector access to cascade halls

Peer reviewed section

^{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 nonproliferation 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% (counting statistical reproducibility) and an accuracy of 0.3% (includes systematic error) 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 ^{3}He 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 up to 1.8 m on a side resulting in detector volumes that are large. The typical well counters have diameters in the range of 10 to 25 cm.
- **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. It should be noted that 3He detectors have been designed with more than one row of tubes to increase the efficiency. However, the resulting efficiency increase is at the expense of increased cost and increased 3He use. The second row of tubes provides about ½ the efficiency per tube as the first row for typical designs. The most efficient use of the 3He gas is with a single row of tubes.

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.

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.

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 the 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.

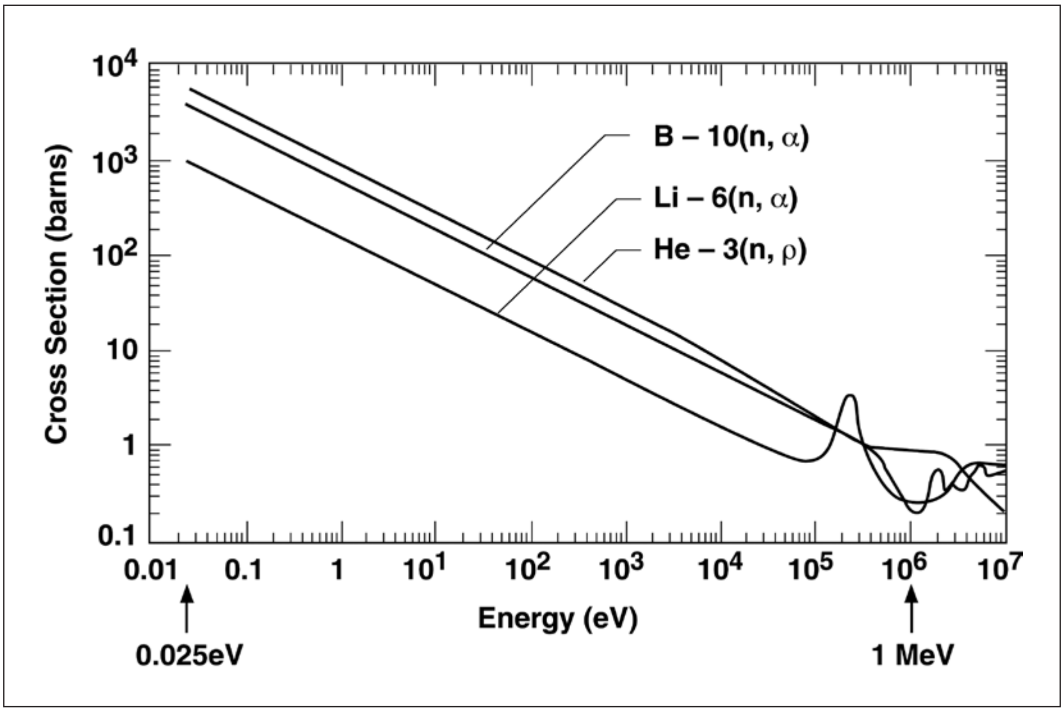

Figure 1: Neutron cross sections as a function of energy for the leading capture reactions being considered for ^{3}He replacement.

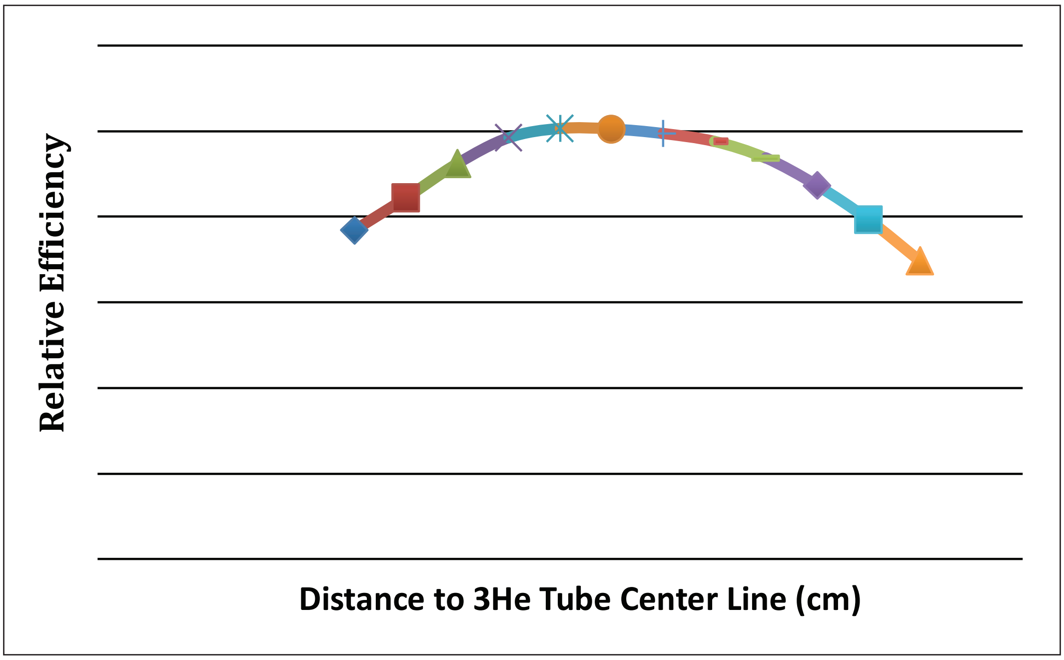

Figure 2: Relative efficiency from MCNPX of a ^{3}He tube (1 inch diameter and 4 atm pressure) embedded in a 11-cm-thick slab of HDPE as a function of depth into the HDPE for fission energy spectrum neutrons.

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.

BF_3 Tubes Considerations – 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.

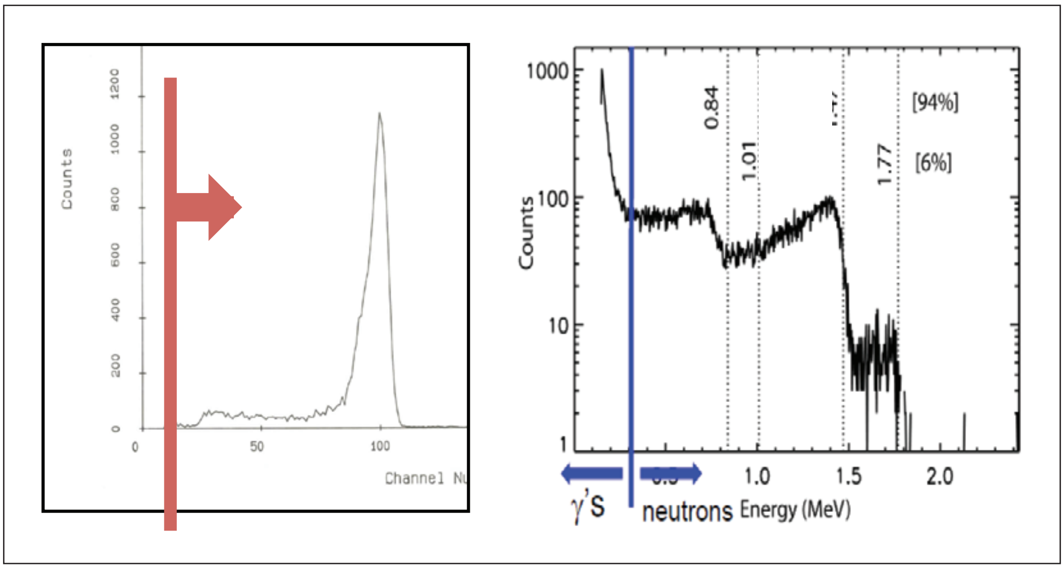

Figure 3: Pulse shape distributions for a ^{3}He tube with 2 μs shaping times (left side) and a thin ^{10}B deposit in an Ar plus methane filled gas tube (right side).

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.

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 de-

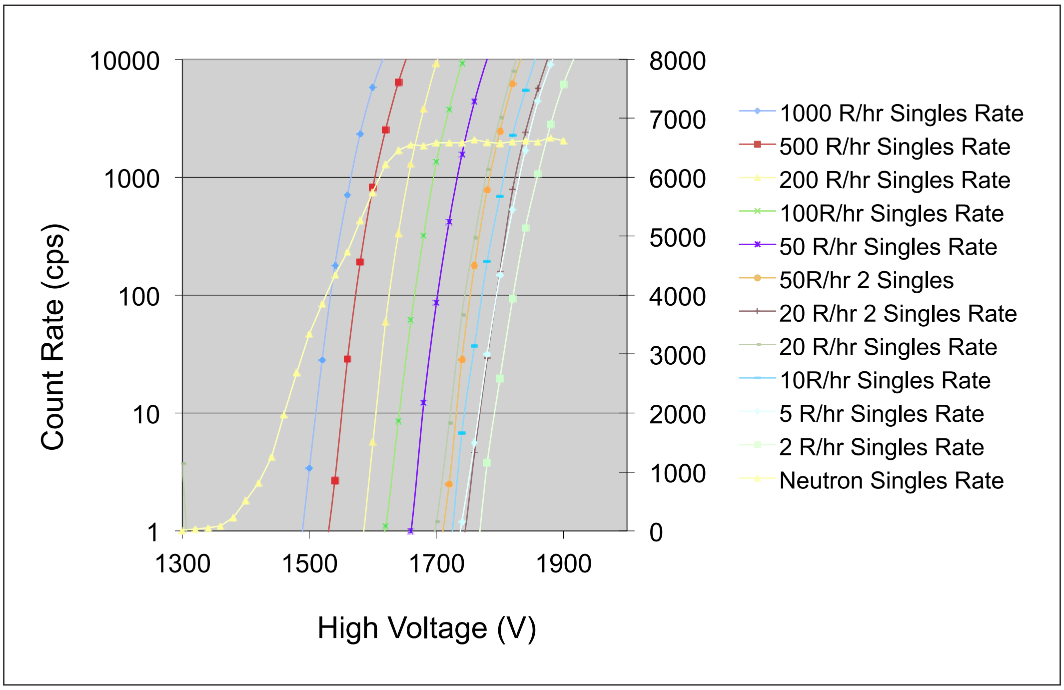

Figure 4: Gamma-ray pileup measurements as a function of high voltage bias for a 25-mm-diameter, 254-mm-long ^{3}He tube at 4 atm pressure.

tector 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 square root 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.

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

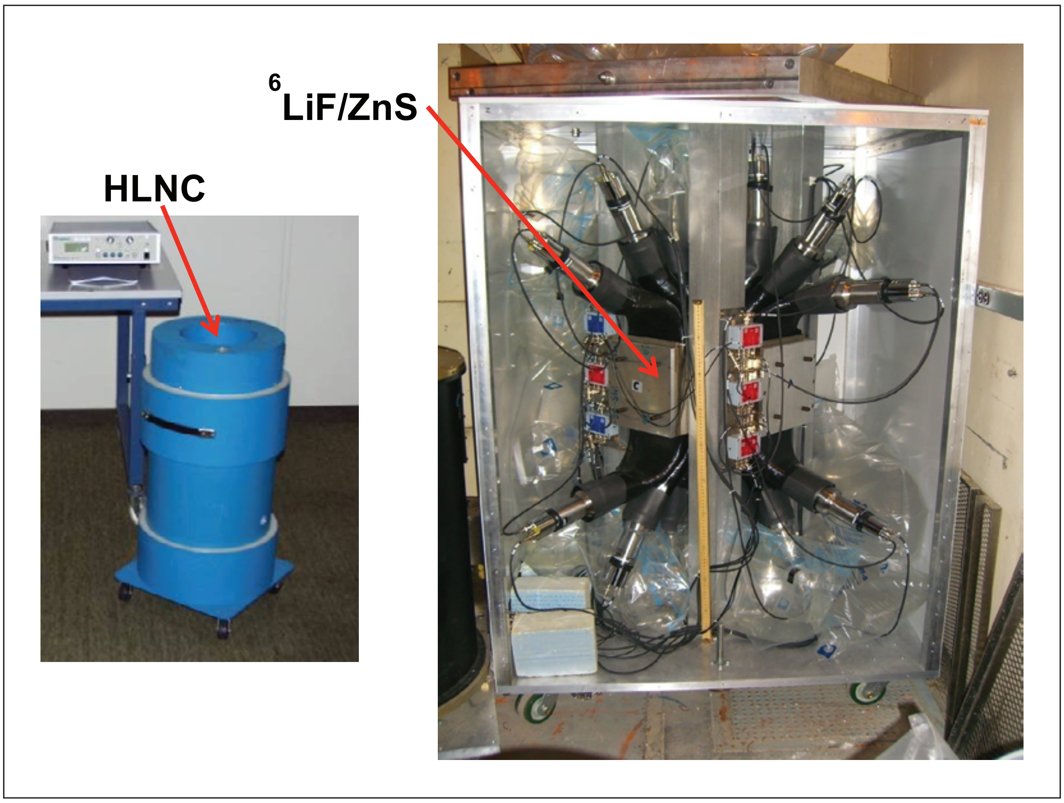

Figure 5: The compact ^{3}He based HLNC Detector (left side), and ^{6}LiF/ZnS scintillator based system (right side) to illustrate the scalability problem.

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

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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 an enhanced version of MCNPX, more powerful computing resources, and 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, additional data points were generated for the assessment of spatial dependencies, spatial plutonium distribution, and the dependence upon fuel shuffling schemes (core loading patterns).

In addition to efforts invested in the characterization of plutonium in the various shuffling schemes, 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

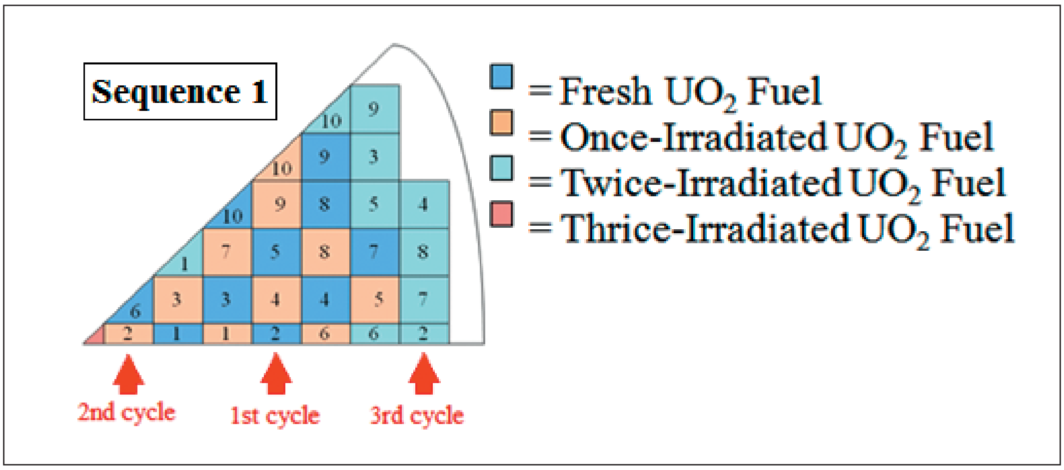

Figure 1: Fuel Shuffling Sequence 1 – Fuel Bundle #2

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 in SFL2 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.

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, this shuffling sequence follows traditional fuel loading practices which keeps the bundle in a central location early in life, and rotates to the core periphery late in life as the bundle reactivity is dropping. 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.

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 2c. Seqence 3 shows a very atypical loading sequence, where fresh fuel is placed on the core periphery. While this would not be performed from a cycle optimization standpoint, it creates steep gradients across the bundle

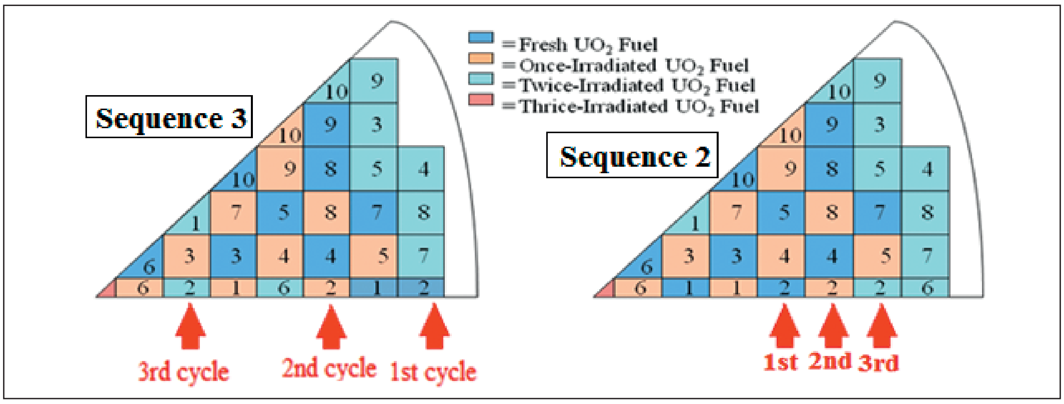

Figure 2: Shuffling Sequence 3 and 2 – Fuel Bundle #2

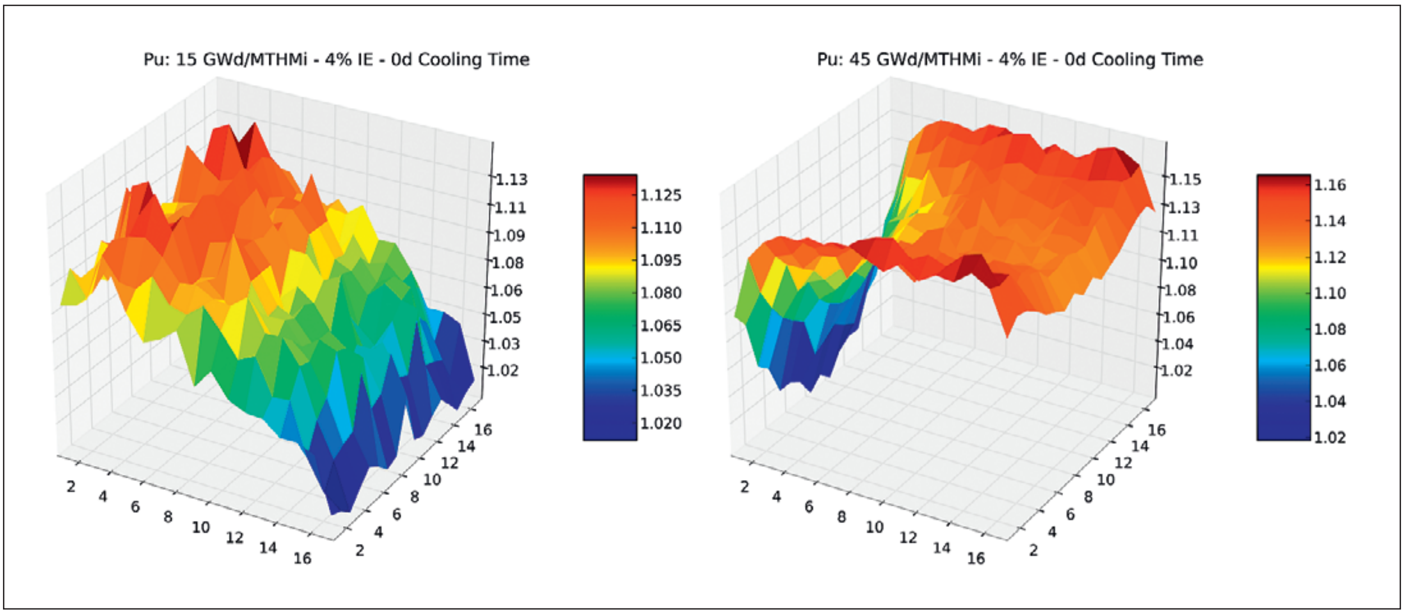

Figure 3: Shuffling Sequence 1 Pu Distribution – 15, 45 GWd/MTU

useful for assessing the impact of strong gradients. 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.

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. 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.

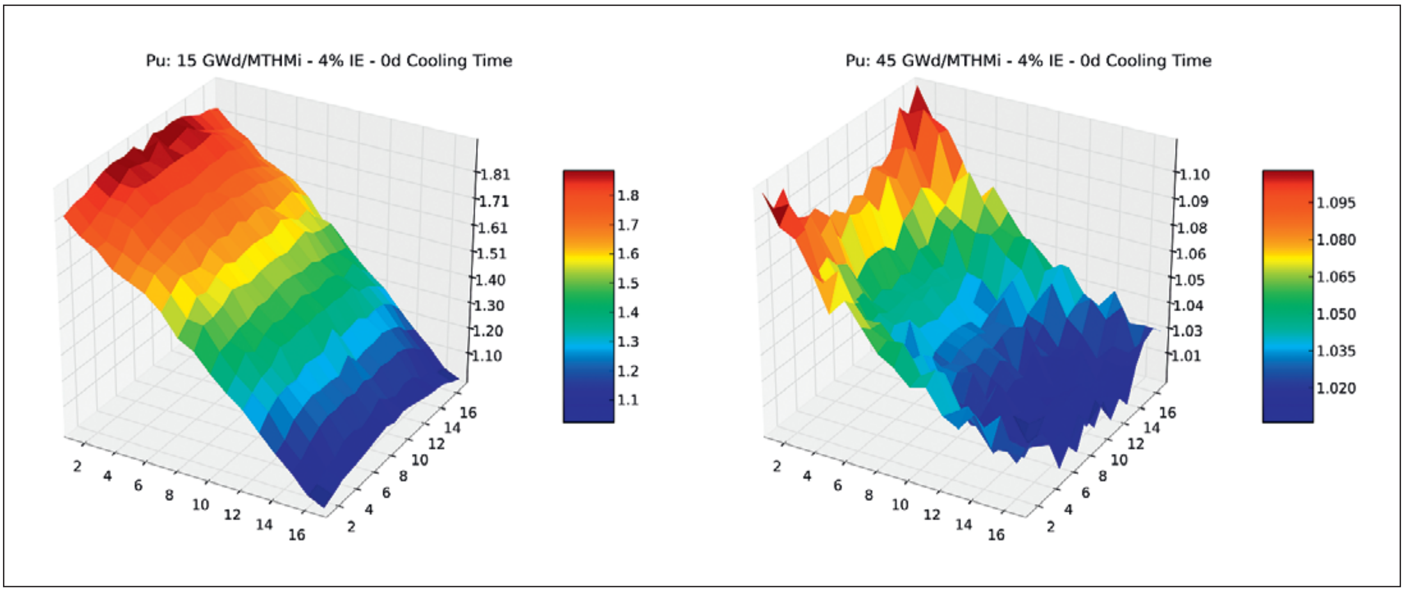

Figure 4: Shuffling Sequence 3 Pu Distribution – 15, 45 GWd/MTU

3.1. Assembly 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 two.

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.

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 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)

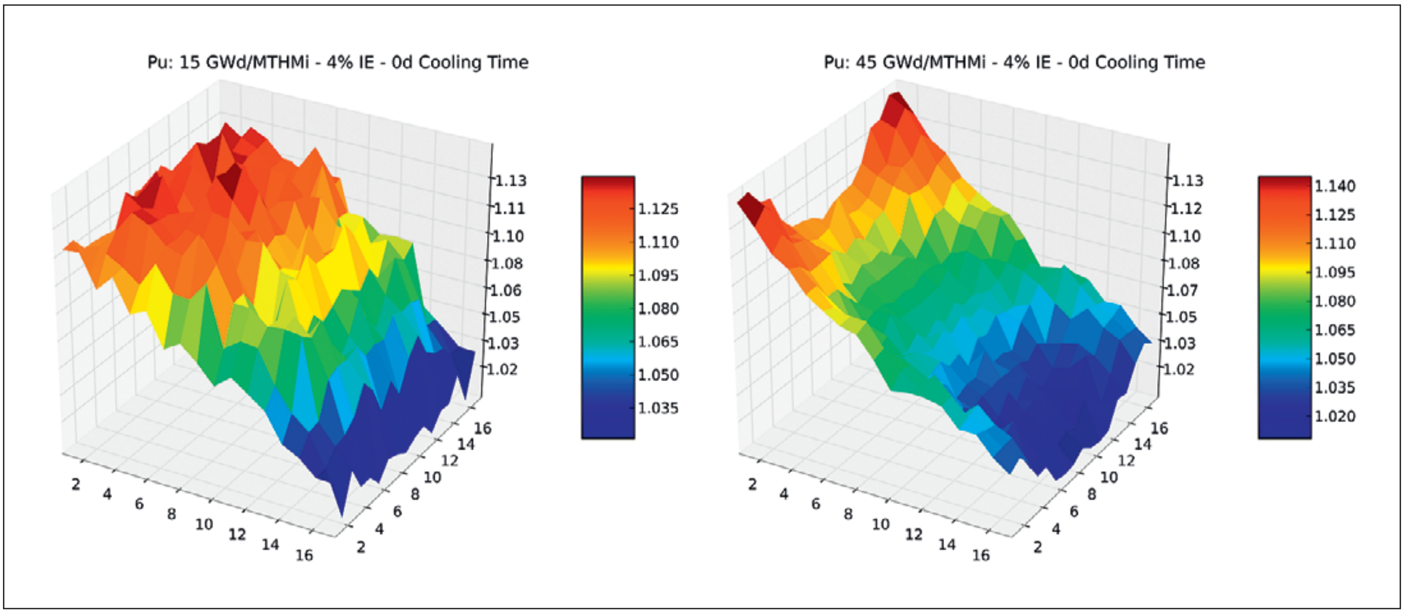

Figure 5: Shuffling Sequence 2 Pu Distribution – 15, 45 GWd/MTU

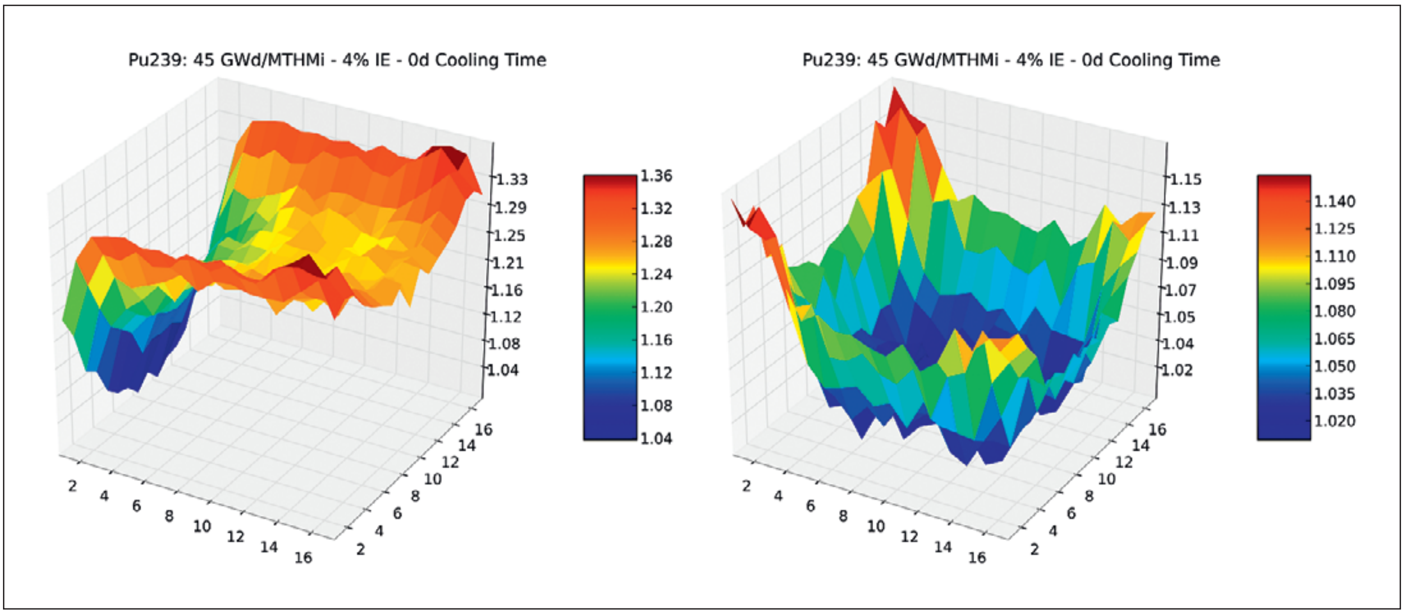

Figure 6: ^{239}Pu Concentration – Shuffling Sequence 1 (left), and Shuffling Sequence 3 (right)
Both Images at 45 GWd/MTU

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.

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.

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%	-0.82%
% Pu239 Change from Sequence 1		4.19%	3.45%

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%	-0.82%
% Pu239 Change from Sequence 1		4.19%	3.45%

Table 2: ^{239}Pu – Elemental Pu and ^{239}Pu Comparison

Lastly, seen in

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%	-0.82%
% Pu239 Change from Sequence 1		4.19%	3.45%

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. As for sequence 3, the ^{239}Pu difference was still greater than sequence 1, being 3.45% but the overall Pu inventory was less being -0.82% less. 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.

3.2. Spatial Plutonium Distribution Prediction

Since the Pu distribution is preferentially weighted toward the bundle periphery, a scheme was developed that utilizes the periphery Pu concentration to predict the Pu concentration for each internal fuel pin location. Using the results from SFL 1, the infinitely reflected assembly, a scheme to predict the Pu concentration for every internal pin was generated, assuming that the Pu quantity was known for each of the fuel pins on the bundle edge. Figure 7 illustrates process flow. Figure 7a represents the pin-by-pin predictor values, computed according to Equation 1. This example uses the four edge Pu concentrations for row and column 7, as well as the known mass at location (7,7). Once these predictors (p[i,j]) have been computed for all internal locations, these predictors are then used to predict what the Pu concentration at every [i,j] location would be ($\tilde{P}[i,j]$). In our example here the $\tilde{P}(7,7)$ location would be the predicted Pu mass for one of the shuffling schemes described above; again assuming that the Pu concentrations in the edge fuel pins is known. Once this is performed for all [i,j] internal locations, a comparison of the known values to the predicted values is then carried out.

$$Predictor_{i,j} = \frac{Pu_{i,j}}{\left[Pu_{j,0} * \frac{(xl-i)}{xl} + Pu_{j,xl} * \frac{i}{xl} + Pu_{0,i} * \frac{(yl-j)}{yl} + Pu_{yl,i} * \frac{j}{yl}\right]}$$

$$xl = number\ of\ pins\ x\ direction;\ yl = number\ of\ pins\ in\ y\ direction$$

Equation 1: Spatial Pu prediction equation

Applying this scheme to the first and third shuffling sequences yields the results in Figure 8 and Figure 9. The error associated with such a prediction scheme is clearly dependent upon the shuffling sequence the fuel bundle experienced. For the first shuffling sequence the error gen-

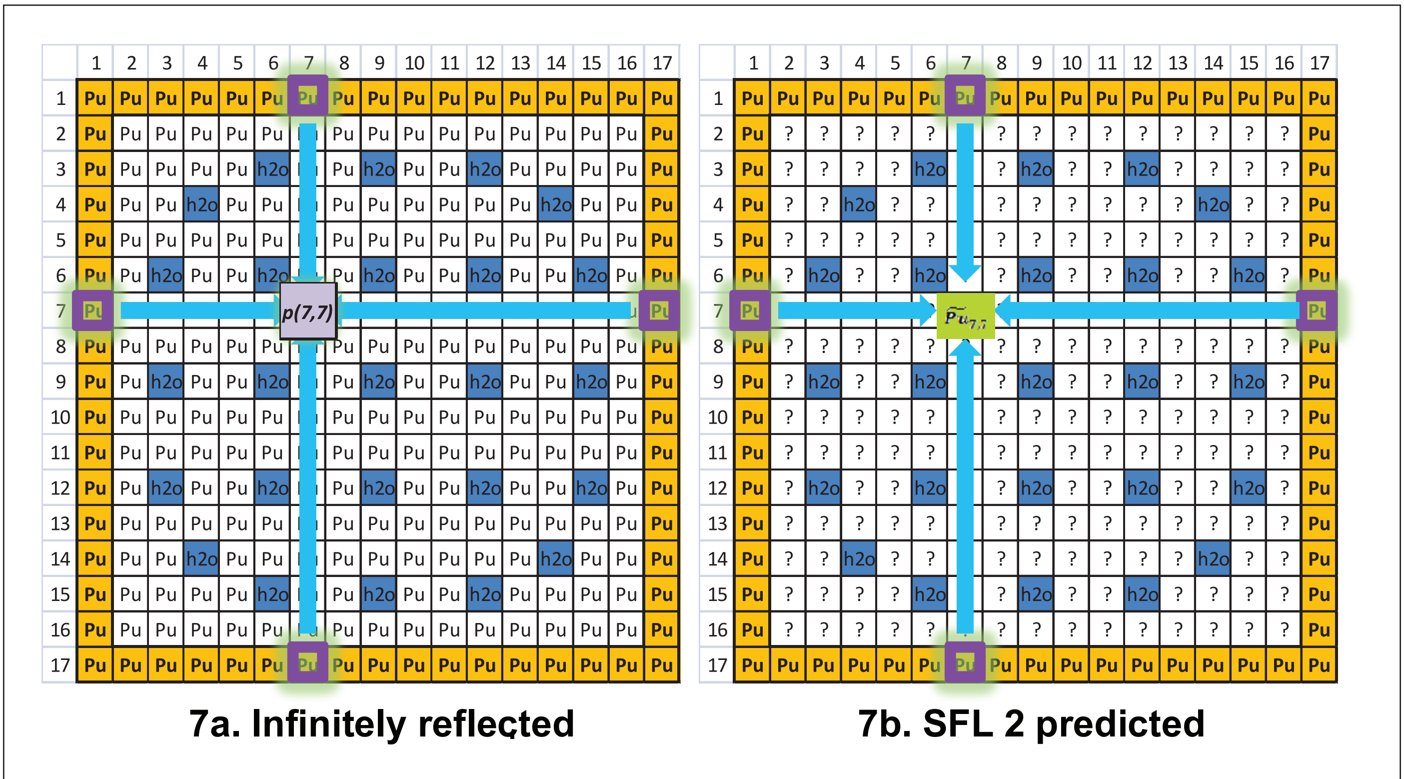

Figure 7: Spatial Pu prediction scheme

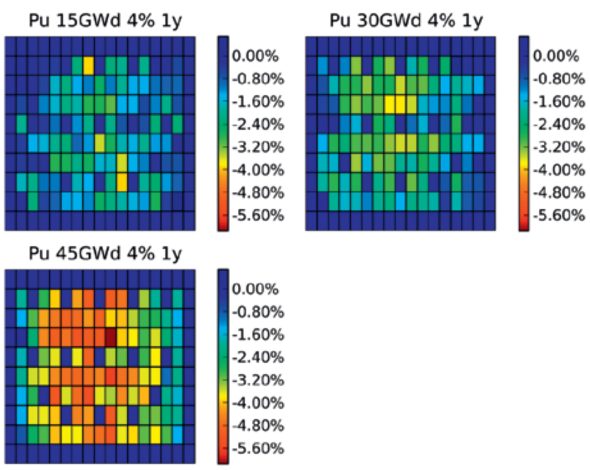

Figure 8: Sequence 1 Pu spatial error – 4% IE, 1y CT

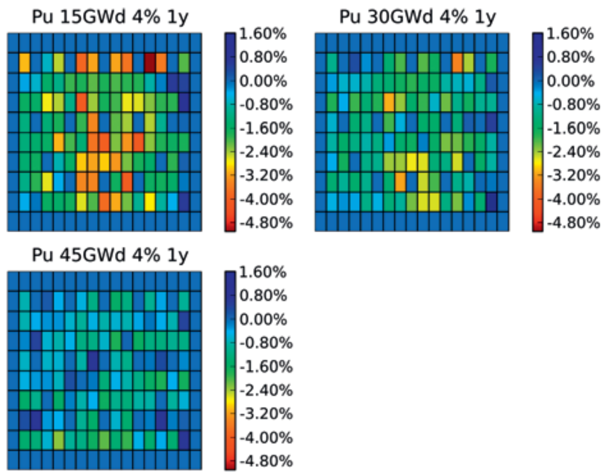

Figure 9: Sequence 3 Pu spatial error – 4% IE, 1y CT

erally increased from a maximum of ~2.5% at 15 GWd/MTU, to a maximum error near 6% at 45 GWd/MTU. In contrast, the second shuffling sequence encountered the greatest error of ~5% at 15 GWd/MTU, while the smallest error of ~1.5% was encountered at 45 GWd/MTU. These results are due to usage of the infinitely reflected assembly to generate the internal predictors. The first shuffling sequence encountered the strongest asymmetries across the bundle at 45 GWd/MTU, observed in Figure 3, which corresponded to the greatest errors. The third shuffling sequence experienced the greatest asymmetries at 15 GWd/MTU, seen in Figure 4, which corresponded to the greatest errors in Pu prediction. This result is expected since a Pu distribution derived from an infinitely reflected assembly was used to predict an asymmetrically burned distribution with no adjustments made for the differing spatial Pu gradients between the base case and the shuffled bundle.

The development of a scheme that would use not only perimeter Pu concentrations, but which would also account for the magnitude of the difference between the peripheral concentrations for a given row or column would be expected to offer a better prediction. Here sequences one and three are shown since they offered the most interesting cases, as well as served as the bounding error cases.

3.3. Assembly Pu prediction

In lieu of predicting pin-by-pin Pu isotopics, a second approach involved the prediction of the total Pu concentration in the interior of the bundle. This approach was an investigation of the benefit encountered in trading spatial fidelity in favor of potential Pu accuracy. In this approach the total mass of Pu located on the bundle periphery was summed and divided by the total mass of Pu in the interior pins of the bundle and called the Pu_{ratio}, seen in Figure 10. Again

$$Pu_{ratio\ SFL\ 1} = \frac{\sum x_{SFL\ 1}}{\sum y_{SFL\ 1}}$$

$$\sum y_{SFL\ 2} = \left[\frac{\sum x_{SFL\ 2}}{Pu_{ratio\ SFL\ 1}}\right]$$

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
1	x	x	x	x	x	x	x	x	x	x	x	x	x	x	x	x	x
2	x	y	y	y	y	y	y	y	y	y	y	y	y	y	y	y	x
3	x	y	y	y	y	h2o	y	y	h2o	y	y	h2o	y	y	y	y	x
4	x	y	y	h2o	y	y	y	y	y	y	y	y	y	h2o	y	y	x
5	x	y	y	y	y	y	y	y	y	y	y	y	y	y	y	y	x
6	x	y	h2o	y	y	h2o	y	y	h2o	y	y	h2o	y	y	h2o	y	x
7	x	y	y	y	y	y	y	y	y	y	y	y	y	y	y	y	x
8	x	y	y	y	y	y	y	y	y	y	y	y	y	y	y	y	x
9	x	y	h2o	y	y	h2o	y	y	h2o	y	y	h2o	y	y	h2o	y	x
10	x	y	y	y	y	y	y	y	y	y	y	y	y	y	y	y	x
11	x	y	y	y	y	y	y	y	y	y	y	y	y	y	y	y	x
12	x	y	h2o	y	y	h2o	y	y	h2o	y	y	h2o	y	y	h2o	y	x
13	x	y	y	y	y	y	y	y	y	y	y	y	y	y	y	y	x
14	x	y	y	h2o	y	y	y	y	y	y	y	y	y	h2o	y	y	x
15	x	y	y	y	y	h2o	y	y	h2o	y	y	h2o	y	y	y	y	x
16	x	y	y	y	y	y	y	y	y	y	y	y	y	y	y	y	x
17	x	x	x	x	x	x	x	x	x	x	x	x	x	x	x	x	x

Figure 10: Assembly Pu prediction – Pu_{ratio} calculation

		Infinite ratio					
		Shuffling Sequence 1		Shuffling Sequence 2		Shuffling Sequence 3	
		Prediction (g)	% Error	Prediction (g)	% Error	Prediction (g)	% Error
15 GWd/MTU	0d	2029.87	-1.74%	1990.21	-2.10%	1848.49	-3.45%
	5y	2043.02	-1.69%	2004.67	-2.04%	1855.69	-3.45%
	80y	1925.01	-1.34%	1891.61	-1.72%	1742.65	-3.63%
30 GWd/MTU	0d	3079.52	-2.29%	3118.38	-1.70%	2991.93	-1.89%
	5y	3032.16	-2.22%	3065.12	-1.68%	2952.17	-1.89%
	80y	2715.03	-2.20%	2740.98	-1.73%	2647.73	-1.91%
45 GWd/MTU	0d	3677.34	-3.68%	3785.31	-1.41%	3722.93	-1.04%
	5y	3577.47	-3.61%	3689.09	-1.35%	3646.23	-0.96%
	80y	3142.06	-3.71%	3236.61	-1.43%	3203.43	-0.88%

Table 3: Assembly Pu prediction error – infinite assembly

some knowledge of the expected behavior of the bundle is required from a base case to project to additional cases, and as before is first obtained from the infinitely reflected assembly. After computing the Pu_{ratio} for the infinitely reflected assembly (SFL 1), this value is then applied to the various shuffling sequences to calculate the total internal Pu mass knowing only the bundle periphery Pu mass.

Table 3 shows the errors encountered in using the infinite assembly (SFL 1) Pu_{ratio} to predict the internal Pu mass for each of the three shuffling sequences. The same trend observed in the spatial Pu prediction was also observed here, with the highest errors occurring at the statepoints subjected to the greatest maximum to minimum Pu swing; the most asymmetric. Errors on the order of 3.5% were the highest observed, at 15 GWd/MTU for sequence three and 45 GWd/MTU for sequence one, while minimum errors were close to 1%. While there is a deficiency in using the symmetrically burned assembly as a baseline to predict the Pu mass in a shuffled bundle, the error associated with this deficiency does not appear to be prohibitive.

To better estimate the total interior Pu mass, a modified approach to the preceding method was developed. This modified approach uses both the Pu_{ratio} for all SFL cases, as previously defined, as well as knowledge about the ratio of the perimeter fuel pin with the highest Pu concentration to the perimeter fuel pin with the lowest Pu concentration. This approach was employed by computing the Pu_{ratio} and the maximum to minimum ratio for the infinitely reflected bun-

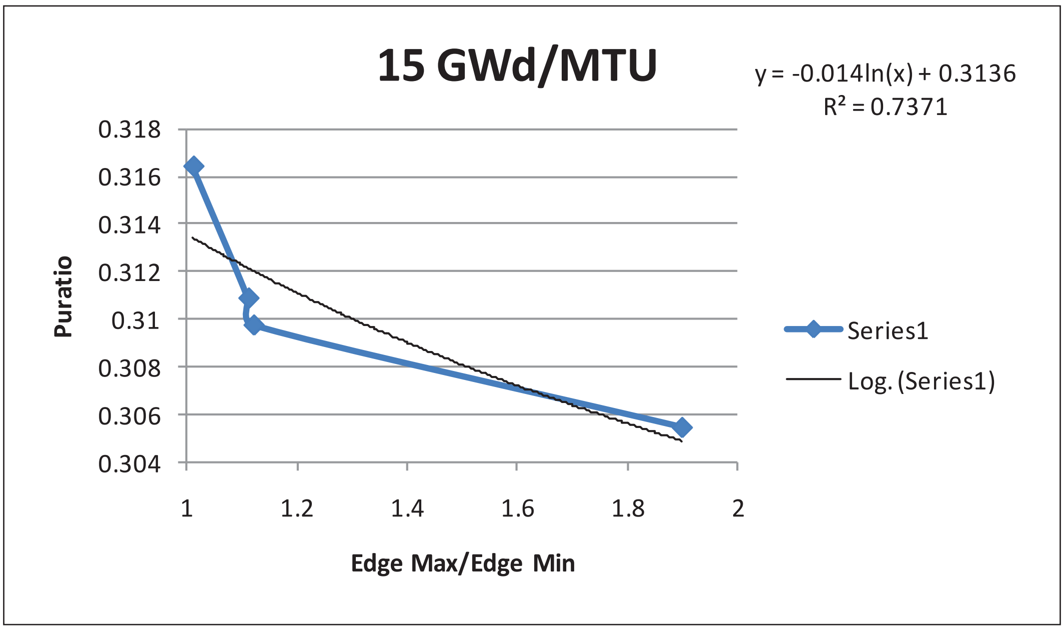

Figure 11: Pu_{ratio} as a function of perimeter max/min ratio

dle, as well as for the three shuffling sequences in the second SFL, plotting the Pu_{ratio} as a function of the maximum to minimum ratio, and fitting a function to the data. Figure 11 shows this data, as well as the fitted function in the form of a logarithmic function for 15 GWd/MTU, where the same approach was applied for 30 and 45 GWd/MTU as well. Here the 15 GWd/MTU results are shown since they include the most extreme maximum-to-minimum pin swing corresponding to the third shuffling sequence in SFL 2.

Using the function obtained in Figure 11, the predicted interior Pu mass was computed for the three shuffling sequences at 15, 30 and 45 GWd/MTU and given here in Table 4. Here the maximum error has been reduced from ~3.5% when using only the Pu_{ratio} from the infinitely reflected assembly, to 1% when treating the Pu_{ratio} as a function of the maximum-to-minimum peripheral pin ratio. In addition to decreasing the prediction error, the trend of the greatest error corresponding to the statepoints with the steepest Pu gradients across the bundle was addressed. The third shuffling sequence intentionally experienced the strongest gradient (via the shuffling sequence employed) at 15 GWd/MTU, however the greatest error was not at this statepoint but rather higher at both 30 and 45 GWd/MTU.

While the results are clearly better when utilizing the functional dependence of the Pu_{ratio} on the maximum-to-minimum swing, this needs further validation since the mass of Pu in the interior pins for the three shuffling sequences was used to calculate the Pu_{ratio}; required for the functional fit. Thus the resulting answers for the three shuffling sequences depended upon knowing the interior Pu distribution beforehand. In order to better address the accuracy in this scheme, additional shuffling sequence simulations need to be run to validate this predictive approach; although the expectation is that the results will be similar to those shown here, especially at 15 GWd/MTU since an extreme gradient condition was created which helps bound the potential problem domain.

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, however in our simulations axial reflection was employed which means that two of the opposing sides yield the same answer and

		Fitted prediction					
		Shuffling Sequence 1		Shuffling Sequence 2		Shuffling Sequence 3	
		Prediction (g)	% Error	Prediction (g)	% Error	Prediction (g)	% Error
15 GWd/MTU	0d	2057.64	**-0.40%**	2018.20	**-0.72%**	1919.86	**0.28%**
	5y	2072.20	**-0.28%**	2034.05	**-0.61%**	1926.60	**0.24%**
	80y	1960.96	**0.50%**	1927.77	**0.16%**	1814.60	**0.35%**
30 GWd/MTU	0d	3119.41	**-1.02%**	3166.21	**-0.19%**	3062.52	**0.43%**
	5y	3073.34	**-0.89%**	3114.20	**-0.10%**	3021.55	**0.42%**
	80y	2765.54	**-0.38%**	2796.61	**0.27%**	2719.02	**0.73%**
45 GWd/MTU	0d	3776.43	**-1.08%**	3874.21	**0.91%**	3778.17	**0.43%**
	5y	3668.65	**-1.15%**	3773.79	**0.91%**	3696.08	**0.39%**
	80y	3225.35	**-1.16%**	3303.94	**0.62%**	3247.23	**0.48%**

Table 4: Assembly Pu prediction error – fitted function

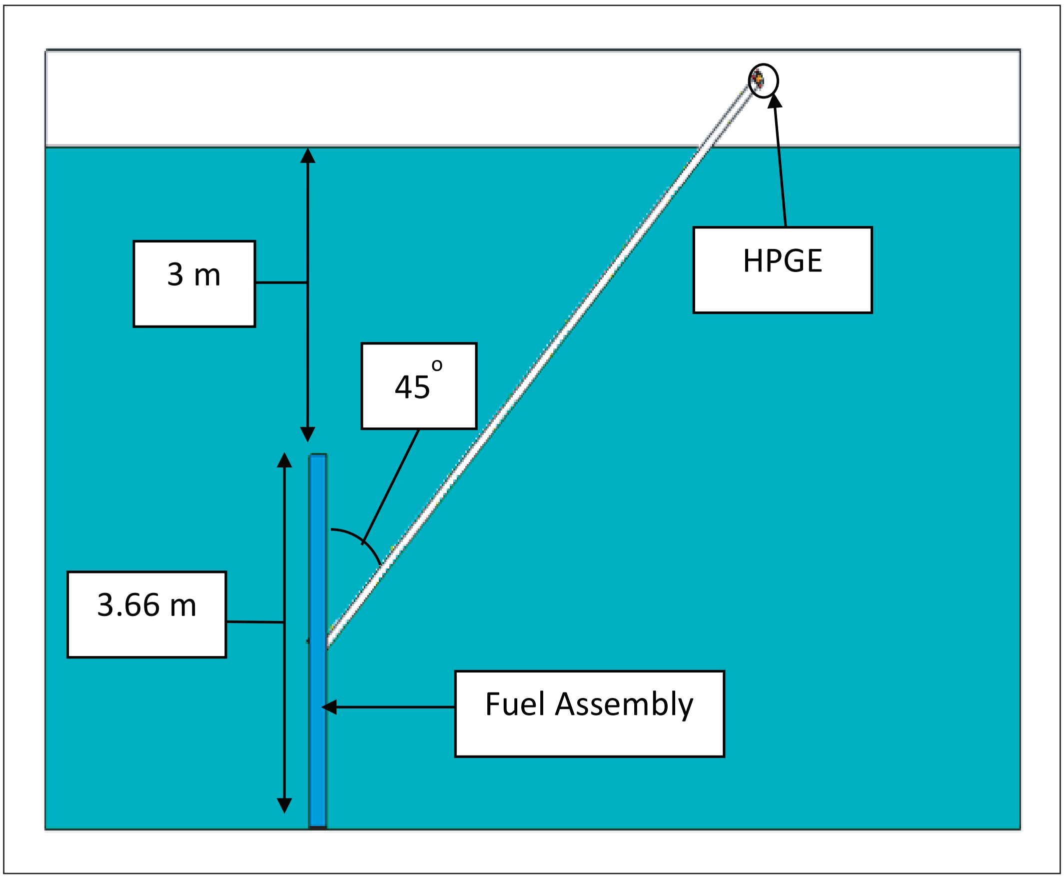

Figure 12: Passive gamma geometry

only three sided simulations were needed. In SFL 2, 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 12 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 the different shuffling sequences.

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 (corresponding to shuffling sequences 1, 2 and 3 respectively) 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, allowing for a wide suite of fuel rotation conditions which can serve well for assessing the potential for BU and IE determination.

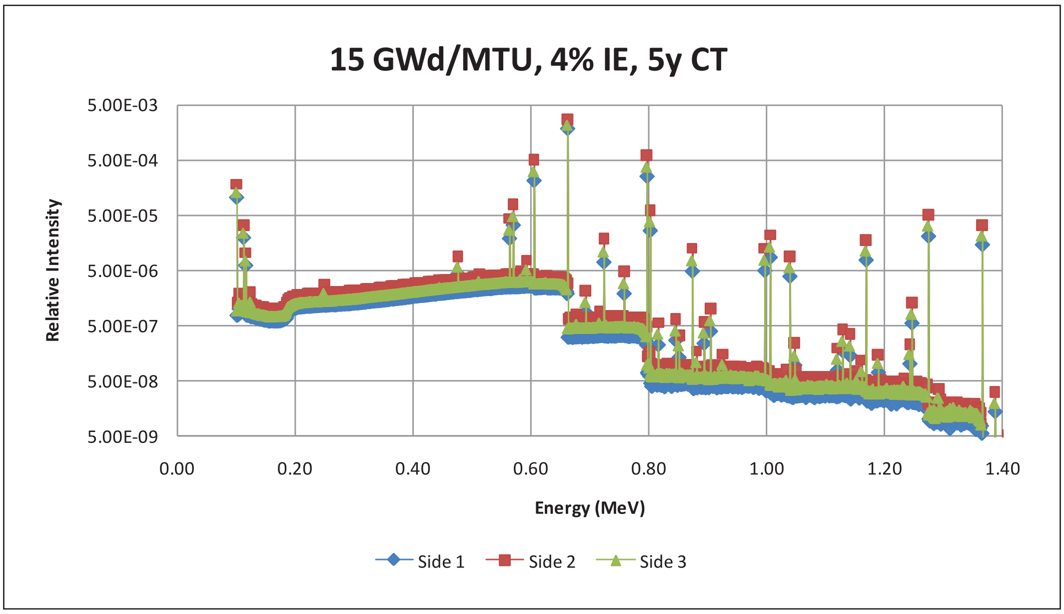

Figure 13: Relative intensity vs. energy (MeV) – 15 GWd/MTU – 5y CT

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 13 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.

Relative difference from side 2 activity (5y cooling time)		
	Side 1	**Side 3**
Cs-134 (.6047 MeV)	-58.84%	-40.86%
Cs-137 (0.6617 MeV)	-32.49%	-23.86%
Cs-134 (0.7959 MeV)	-58.39%	-39.55%

Table 5: Relative difference – 15 GWd/MTU – 5y CT

In quantifiable terms, Table 5 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 5 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 2 and Equation 3 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 5. 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 2: ^{137}Cs production chain

$$133_I \rightarrow 133_{Xe} \rightarrow 133_{Cs(n,\gamma)} 134_{Cs} \rightarrow 134_{Ba}$$

Equation 3: ^{134}Cs production

Figure 14 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 14. 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.

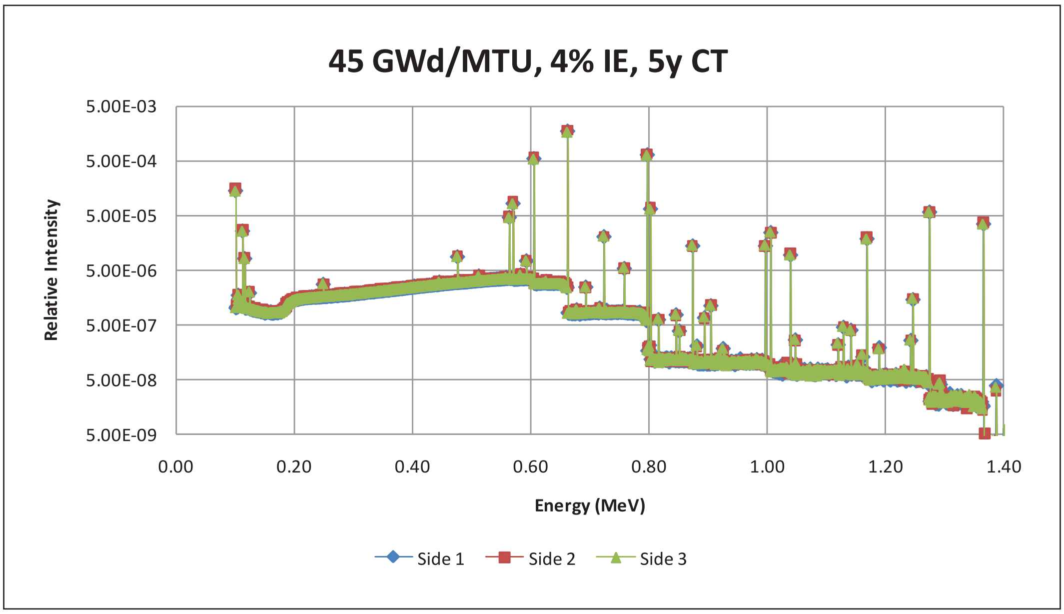

Figure 14: Relative intensity vs. energy (MeV) – 45 GWd/MTU – 1y CT

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 6, 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 6: 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 is rotated from one central location, to a more central location, until 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 and shuffling sequence 1 results at 45 GWd/MTU, Table 7 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

Sequence 3 – 15 GWd/MTU – 5y			
	cs134	cs137	ratio
Side 1	5.43E-04	1.84E-03	**0.2955**
Side 2	1.31E-03	2.72E-03	**0.4801**
Side 3	7.84E-04	2.07E-03	**0.3784**

Sequence 1 – 45 GWd/MTU – 5y			
	cs134	cs137	ratio
Side 1	1.37E-03	1.69E-03	**0.8100**
Side 2	1.46E-03	1.79E-03	**0.8178**
Side 3	1.41E-03	1.71E-03	**0.8258**

Table 7: Cesium Ratios

~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.

5. Conclusions and Future Work

Much work has gone into the generation of SFLs for the NGSI effort 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 preferentially accumulates on bundle edges, and particularly bundle corners. Using known plutonium concentration from the SFLs, predictive schemes for spatial pin-by-pin distribution as well as a bundle total quantity of plutonium from the edge plutonium concentration have been developed. Errors on the order of 3-6% were observed in pin-by-pin distributions, whereas errors of 1-3.5% and potentially less than 1% were seen for assembly interior Pu mass depending upon the predictive scheme utilized. Additional validation of these methods by comparing against additional shuffling schemes would be useful in support of integrated instrument design to the benefit 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 intended to capture the signal that the HPGE detector is exposed to would be beneficial and allow 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 in an attempt to reliably use this simulation technique in combination with a burnup code to predict assembly average BU and potentially additional parameters such as IE or CT.

6. Acknowledgements

The authors would like to acknowledge the support of the Next Generation Safeguards Initiative (NGSI), Office of Nonproliferation and International Security (NIS), and the National Nuclear Security Administration (NNSA) 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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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 that 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 / Pu 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 and plutonium.

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.

Keywords: Instrumentation; LIBS; Safeguards; Applications; Development

1. 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).

2. 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 7 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

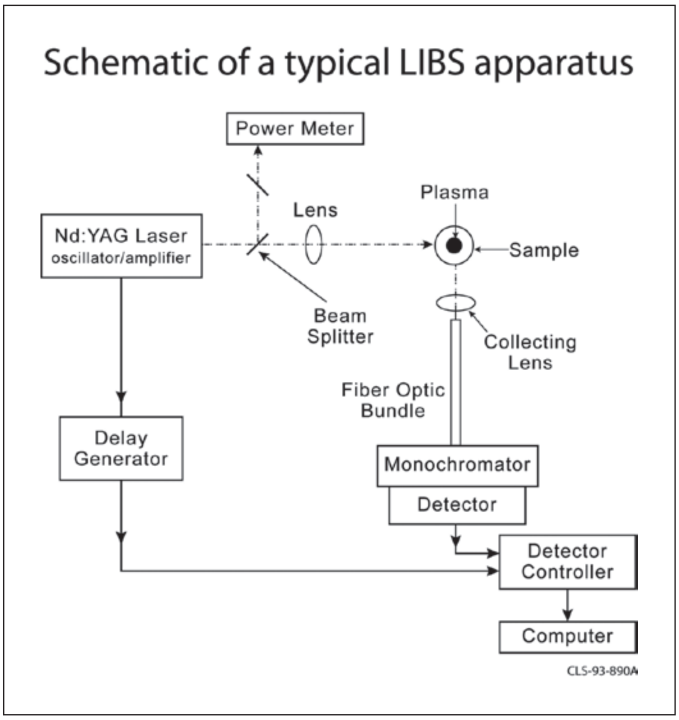

Figure 1: Schematic of a typical LIBS experimental apparatus is shown.

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 in spatial dimension) is generated. Typically, the laser operates at 1064 nanometers with a pulse length of 5 – 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 in a spot approximately 0.5 mm. The emission is collected with a lens and directed to a spectrometer using a fiber optic bundle. The emission is then analyzed by using a computer.

3. Instrumentation Development and Performance Testing

3.1. 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 (Chemistry Division-Chemical Diagnostics and Engineering) 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 equipped with safety interlocks to protect the user. The green enclosure 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.

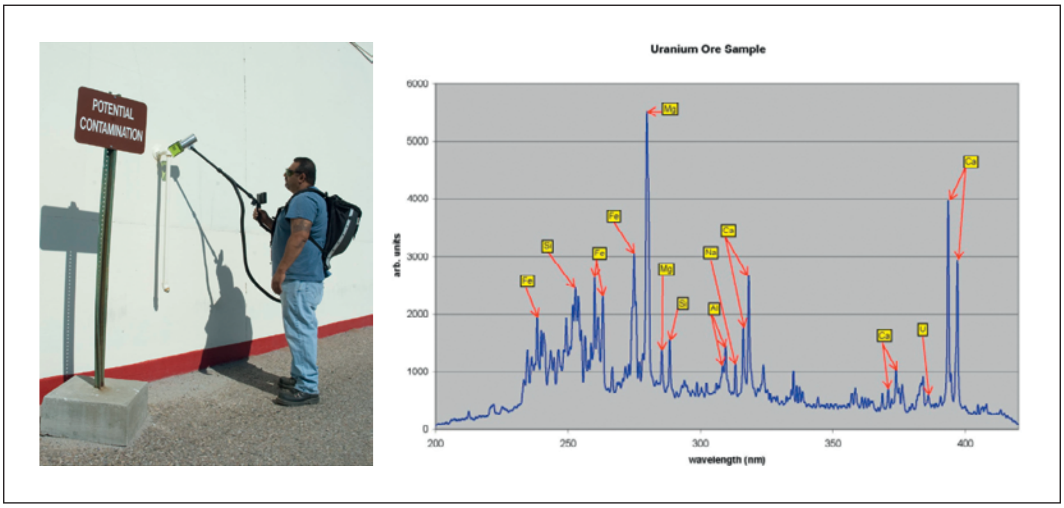

Figure 2: In this picture the backpack system is shown. At the bottom we show a typical LIBS spectrum of a sample of natural abundance uranium ore in the region 200 – 420 nanometers (nm).

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 natural abundance sample of 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. A typical low resolution spectrum of a natural abundance uranium ore sample is shown in Figure 2 between 200 and 420 nanometers. The most intense peaks assigned in the spectrum are not due to uranium atomic emission transitions. The explanation for this observation is twofold at least. First, the density of states for the actinide atoms is very high and the available excitation energy in the plasma must be shared with the high density of states. The results are that the atomic emission transitions in the actinide atoms tend to be weak compared to corresponding atomic emission transition for elements like Ca, Fe, Mg, Na, and Si. Elements that have much simpler electronic configurations and therefore much lower density of excited states that give rise to much simpler atomic emission spectra. The second element of this explanation is that all of the elements in the plasma are excited and again the available excitation energy must be shared by not only the actinide element (uranium) but also the other elements as well. Again, the high density of states, the sharing of available excitation energy, and other quantum physics and photo-dynamic effects like energy transfer and collisional deactivation between excited states lead to the spectral pattern that is shown in Figure 2 for the uranium ore sample. The uranium ore sample or BL-5 is a low grade concentrate from Beaverlodge, Saskatchewan, Canada. The major mineralogical components are, in decreasing order of abundance: plagioclase feldspar ($Na_{65}K_{10}Ca_{25}$), hematite (Fe_2O_3), quartz (SiO_2), calcite ($CaCO_3$), dolemite ($CaMg(CO_3)_2$), chlorite ($(Mg, Fe, Al)_3(Si, Al)_4O_{10}(OH)_2(Mg,Fe,Al)_3(OH)_6$), and muscovite ($KAl_2AlSi_3O_{10}(OH,F)_2$); uranite ($UO_2$) is the main uranium-bearing mineral. The approximate chemical composition of the major elements in this standard sample in weight percent is: Si (22.0), U (7.09), Al (6.0), Fe (5.9), Ca (4.0), Na (3.6), C (1.9), Pb (1.5), Mg (1.5), K (0.4), Ti (0.4), S (0.3), and V (0.1).

By expanding the scale and observing fine structural details for LIBS spectral between 200 and 800 nanometers, 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 respectively. 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.

Wavelength nm	Ionization State	Wave-length 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

Table 1: Uranium peak assignments from low resolution LIBS spectra.

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.

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

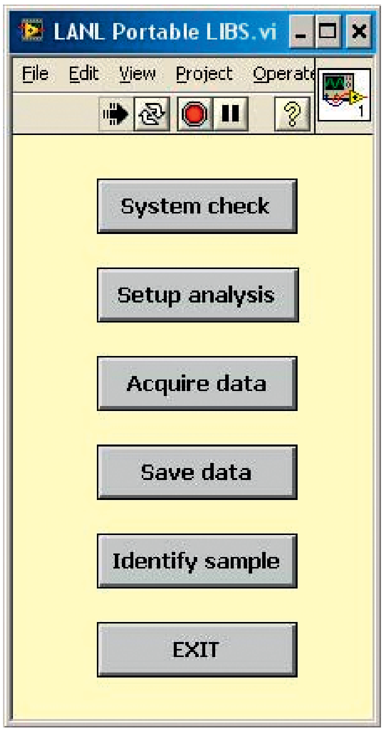

Figure 3: In this figure a transparent user friendly interface for controlling the backpack system and collecting data is shown.

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.

3.2. 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 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. This system also can be used to provide isotopic and ratio analysis of samples of actinides (for example, isotopic measurements on samples

Figure 4: In this picture is shown a general view of the Cart / Rack mounted LIBS system (top view) and the system coupled to a 50 meter fiber optic cable illuminated with a green alignment laser for visual effects (bottom view).

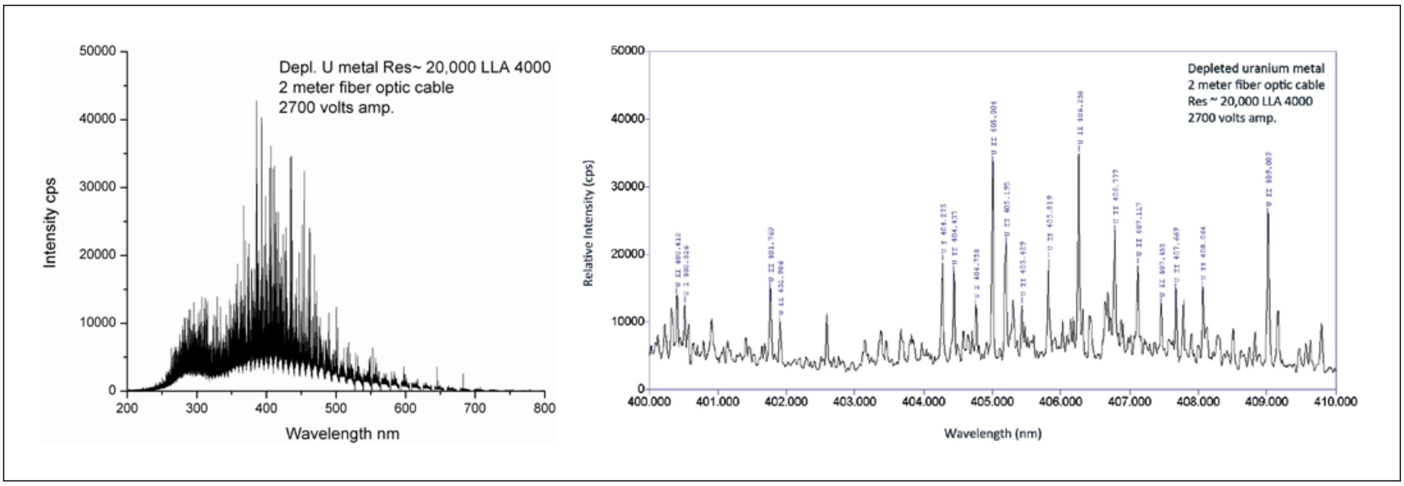

Figure 5: In this figure we show a high resolution spectrum of a sample of depleted uranium between 200 and 780 nanometers. A 10 nm section of the full spectrum is shown at the bottom of the Figure along with peak assignments.

of uranium, and important ratios that include (U / Cm, Pu /, Cm, etc.)).

A prototype version of this system is shown in Figure 4. The top picture 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 optics for collecting 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*** sample 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 located at the bottom 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.

Peak assignments along with relative intensities in counts per second are listed in Table 2. The assigned peak position for the depleted uranium sample are much more accurate for this system since the resolution of the spectrometer is approximately 20,000 compared to 2,000 for the spectrometer used in the backpack mounted portable LIBS system.

Wavelength (nm)	Ionization State	Rel. Int.
400.410	U II	13880
400.526	U I	12509
401.769	U II	14961
401.906	U II	9953
404.275	U I	18649
404.437	U II	17456
404.758	U I	12493
405.004	U II	33641
405.195	U II	21708
405.429	U II	12422
405.819	U II	19184
406.258	U II	34886
406.777	U II	23500
407.117	U II	18307
407.455	U II	12688
407.669	U II	14880
408.064	U II	15144
409.009	U II	25646

Table 2: Uranium peak assignments between 400-410 nm for a depleted uranium metal sample.

A LIBS spectrum of thorium oxide in a stearic acid binder is shown in Figure 6. Stearic acid is an organic binder with the chemical formula $CH_3(CH_2)_{16}CO2H$. The organic binder is necessary to hold the fine power sample of thorium together. The binder does not add to the complexity of the atomic emission spectrum of thorium. Again, the thorium peak assignments are labeled in Figure 6 and listed in Table 3 along with the peak intensities in counts per second.

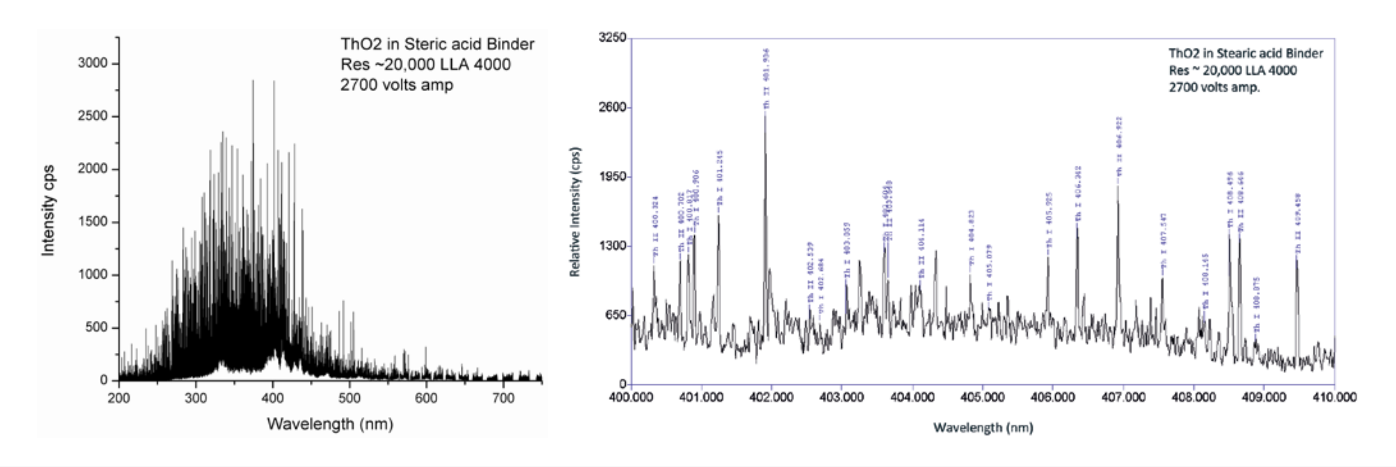

Figure 6: In this figure we show a full spectrum of thorium oxide at the top between 200 and 780 nanometers. An expanded view of a 10 nanometer section is shown at the bottom of the Figure.

Thorium lines for ThO2 in stearic acid.		
Wavelength (nm)	**Ionization State**	**Rel. Int.**
400.324	Th II	1115
400.702	Th II	1161
400.817	Th I	1215
400.906	Th I	1400
401.245	Th I	1588
401.906	Th II	2510
402.539	Th II	703
402.684	Th I	578
403.059	Th I	936
403.604	Th I	1282
403.648	Th II	983
404.114	Th II	880
404.823	Th I	1033
405.079	Th I	725
405.925	Th I	1163
406.342	Th I	1169
406.922	Th II	1865
407.547	Th I	994
408.145	Th I	596
408.496	Th I	1406
408.646	Th II	1365
408.875	Th I	425
409.458	Th II	1159
410.037	Th I	883

Table 3: Thorium peak assignments between 400-410 nm.

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 LIBS system. 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.

3.3. 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 [7]. 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 424.437 nanometer line for uranium was used to perform isotopic analysis on samples of uranium (U-238 / U-235). This measurement was made in our cold LIBS laboratory so the uranium (235) line is not visible in the spectrum shown in Figure 7. However, the uranium (235) line would be observed at 424.412 in an isotopic enriched sample of uranium.

3.4. Single Particle LIBS Microscope Development

Finally, we are also developing a LIBS microscope system that can be used to analyze single particles of samples im-

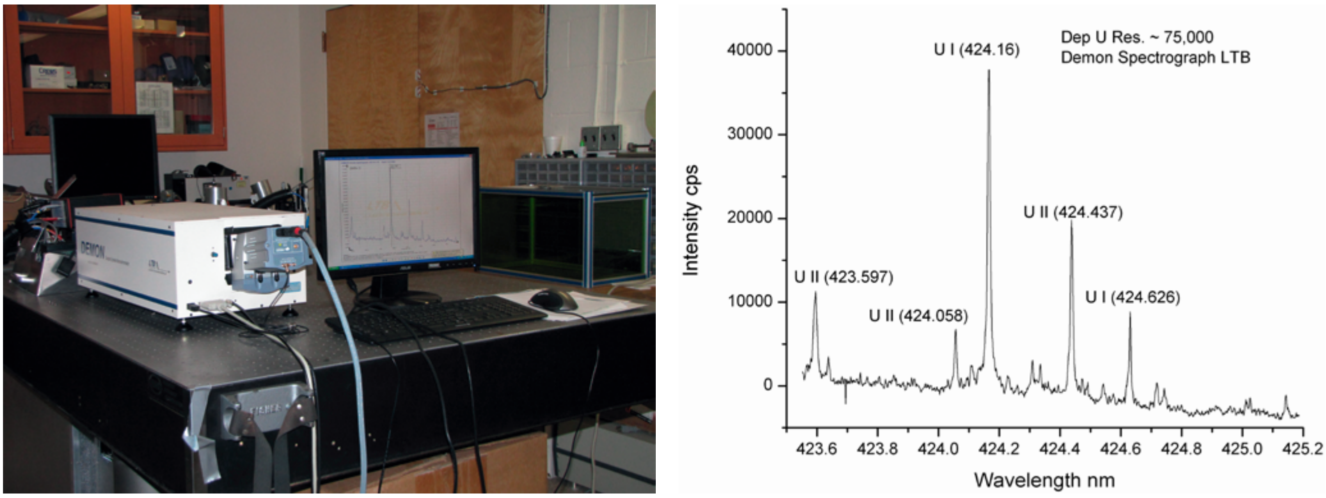

Figure 7: In this figure we show a high resolution spectrograph (75,000). A spectrum of an approximately 2 nanometer section for a depleted uranium metal sample is also shown.

portant 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.

This LIBS microscope can be used to analyze single 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.

We are currently in the process of using this system to analyze single particles of depleted uranium and thorium. The results of this investigation will be the subject of future reports.

4. 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

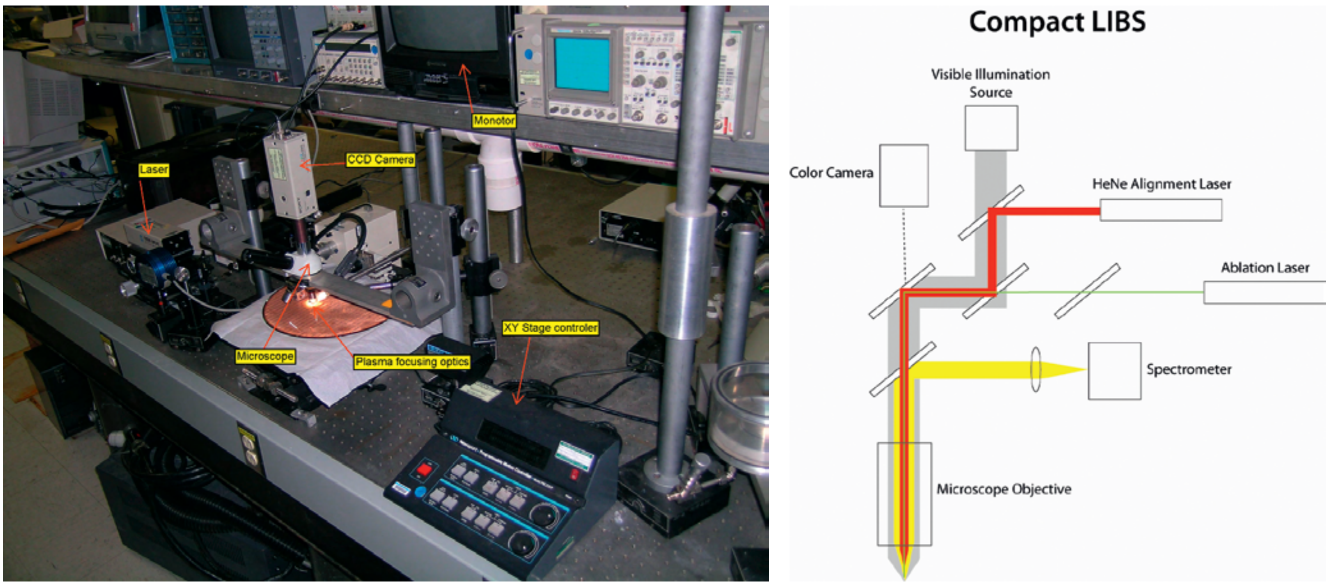

Figure 8: We show an image and a schematic of a compact version of the LIBS microscope in this figure.

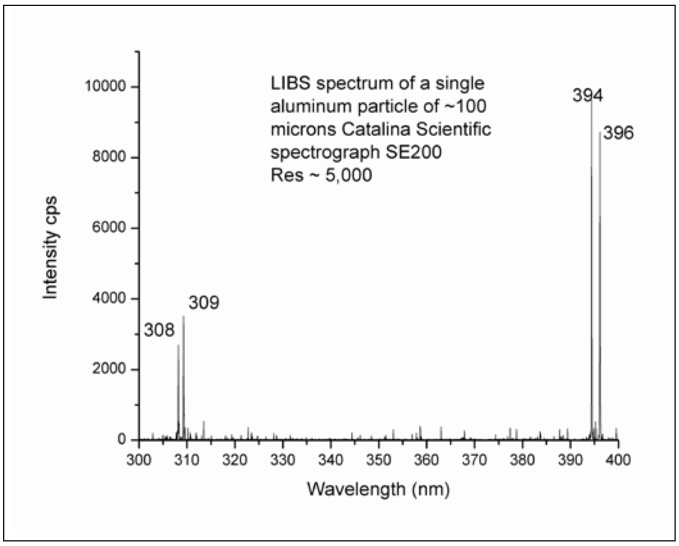

Figure 9: Shown is a LIBS spectrum of an approximately 100 micron particle of aluminum.

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, pre-screening 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.

5. Acknowledgements

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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 (SMMS), comprising data collection instruments, data transmission equipment and an advanced Solution Monitoring Software (SMS), is being implemented at a large scale reprocessing plant in Japan. 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), comprising instruments which provide signals for pressure, temperature and/or neutron counts, the data transmission equipment, and an associated enhanced software (SMS), 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 that includes dissolution/clarification, extraction, purification and concentration, and the High Active Liquid Waste treatment/storage. SMMS involves over 90 vessels or other equipment (evaporators, extractors) 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, e.g: the input/output accountancy tanks, some key vessels between the two extraction cycles and after the 2nd cycle, finally all tanks with highly concentrated Pu solution 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.

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

Figure 1: Results of the automatic evaluation of cycles, solution transfers and solution status

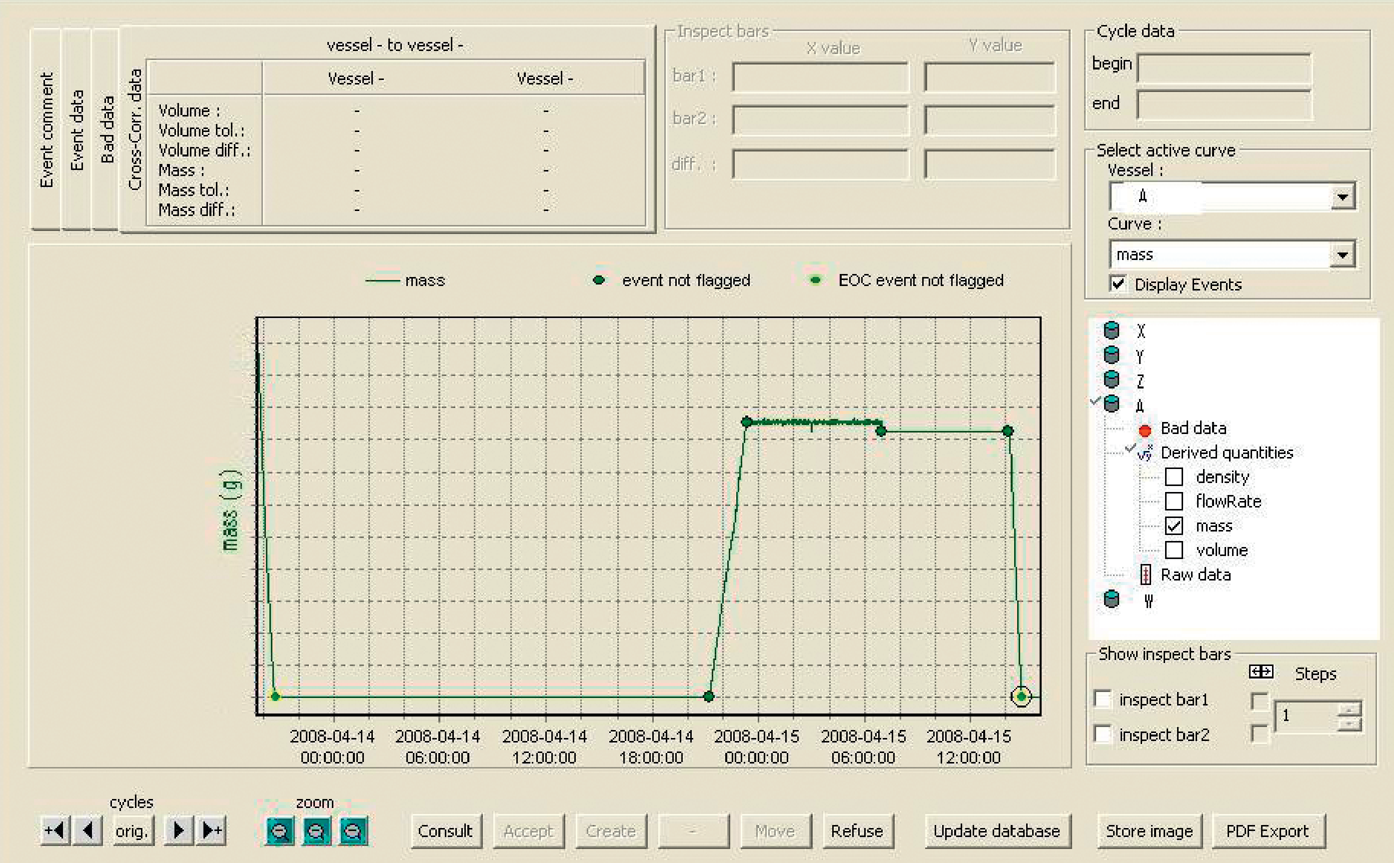


Figure 2: Data Graph display of accountability tank cycle–automatically positioned events (green circle) associated with the detection of functional behaviour

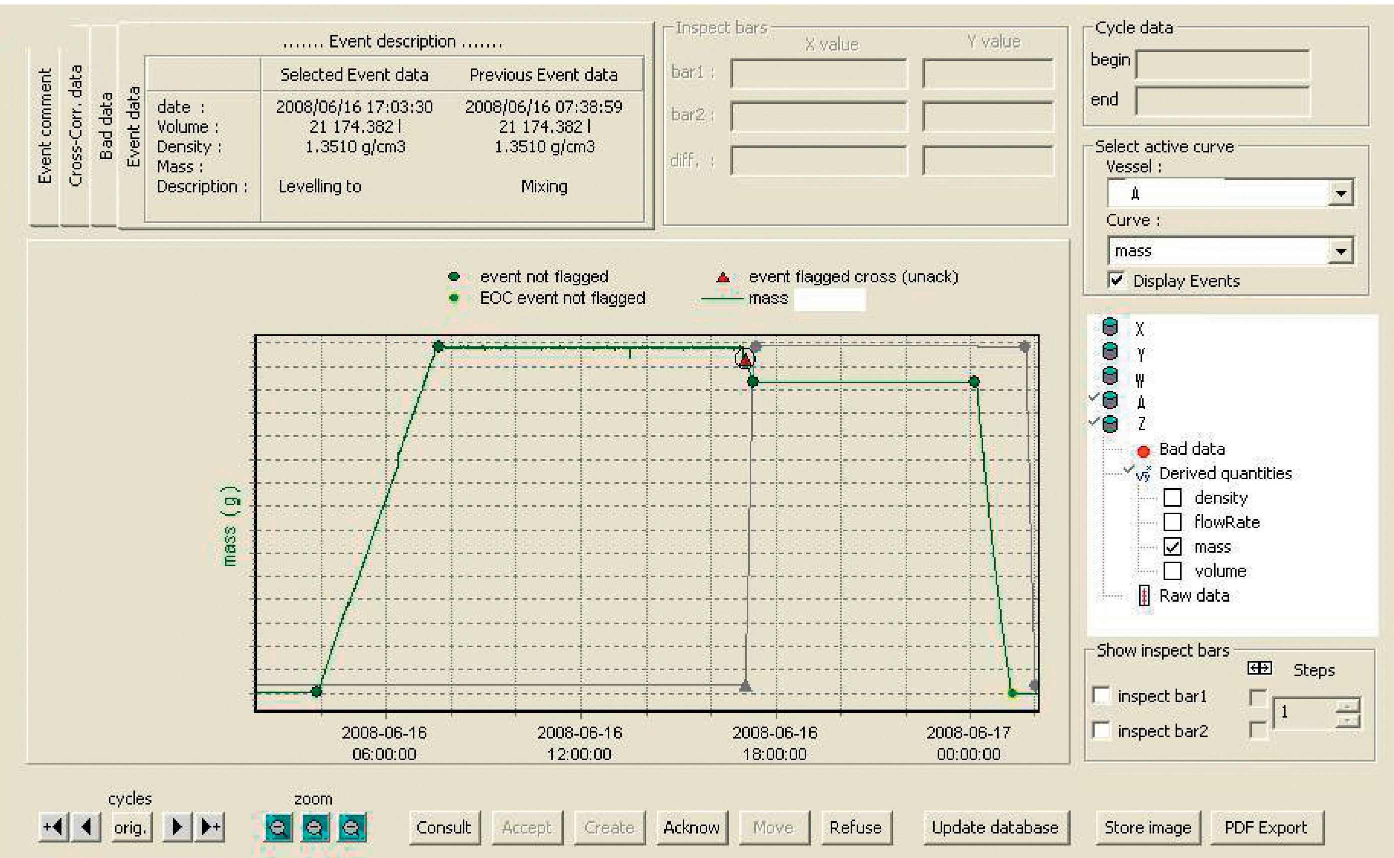


Figure 3: Data display of an accountability tank cycle: a solution transfer is flagged as a cross-correlation alarm

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

Figure 4: Historical Summary of solution transfers between defined partner vessels within a defined time period

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.

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.

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.

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 advan-

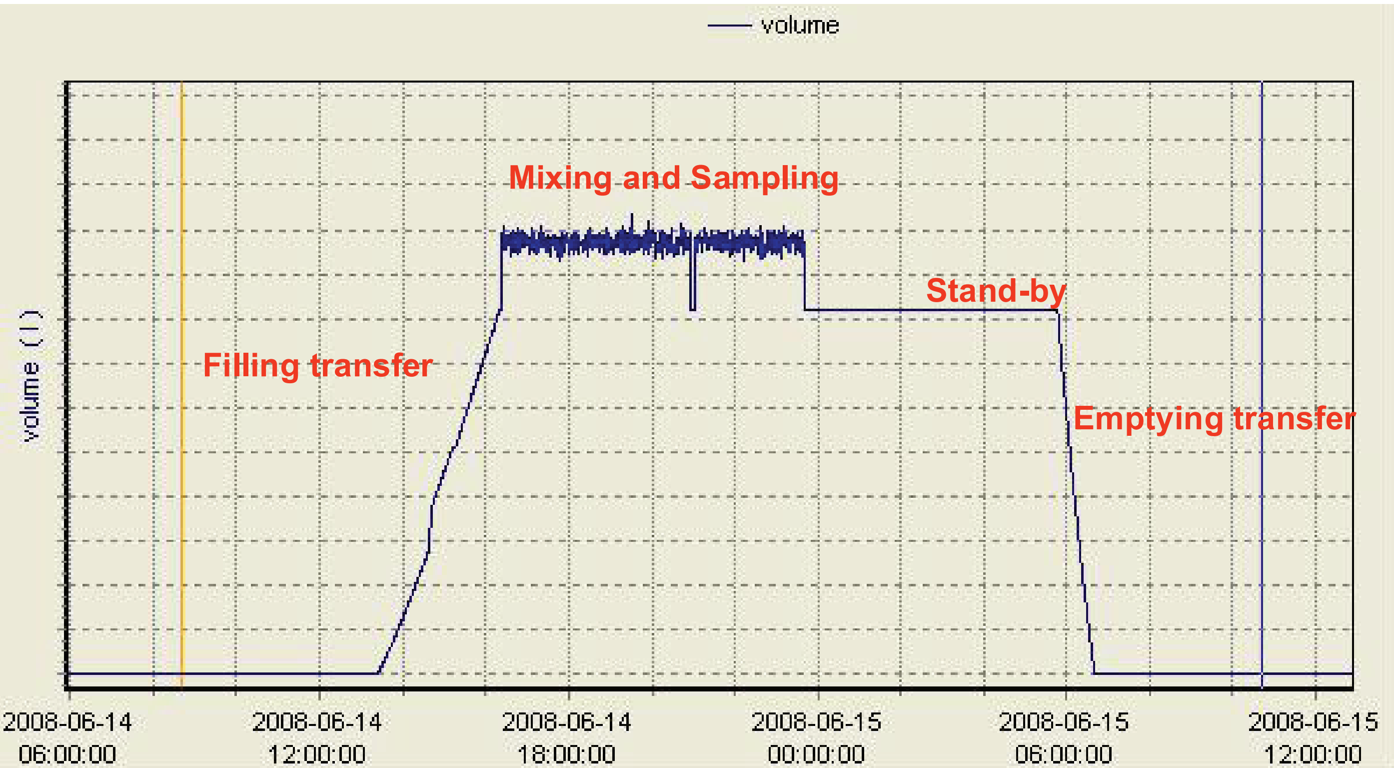


Figure 5: Data display of a typical cycle for an accountability tank

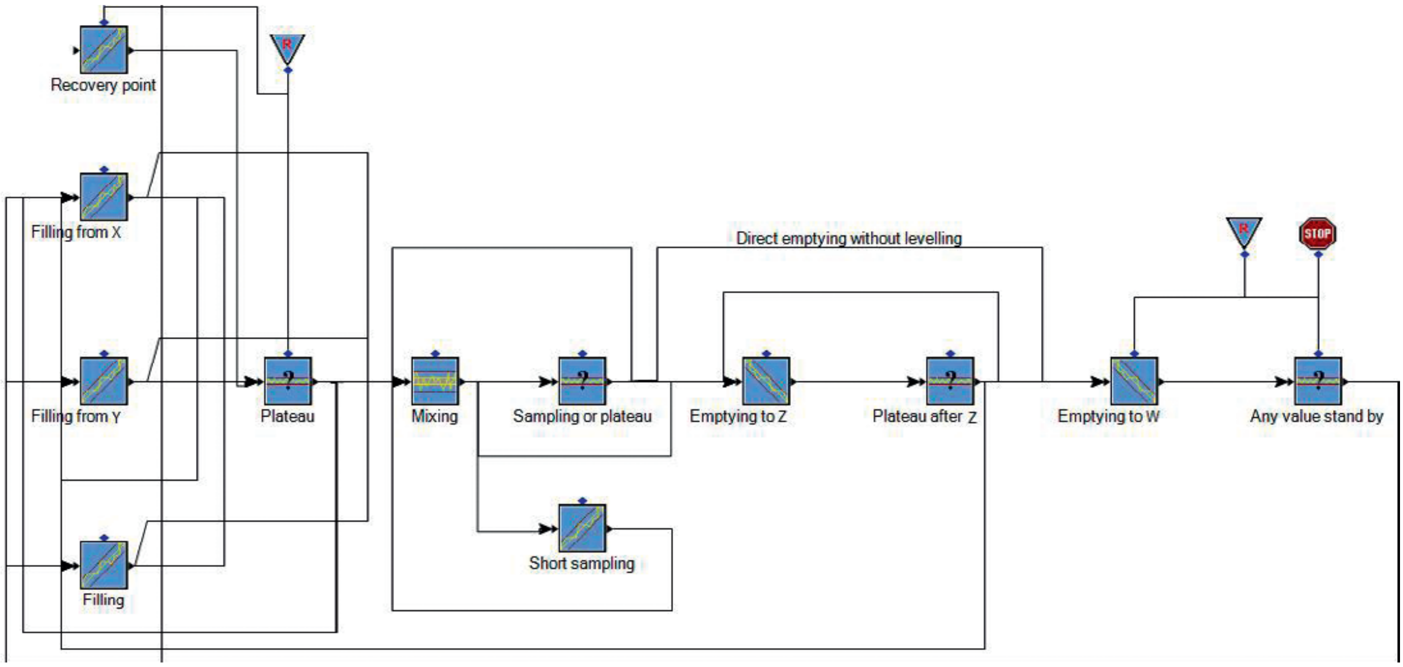


Figure 6: Reference Signature for an accountability tank

tage 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.

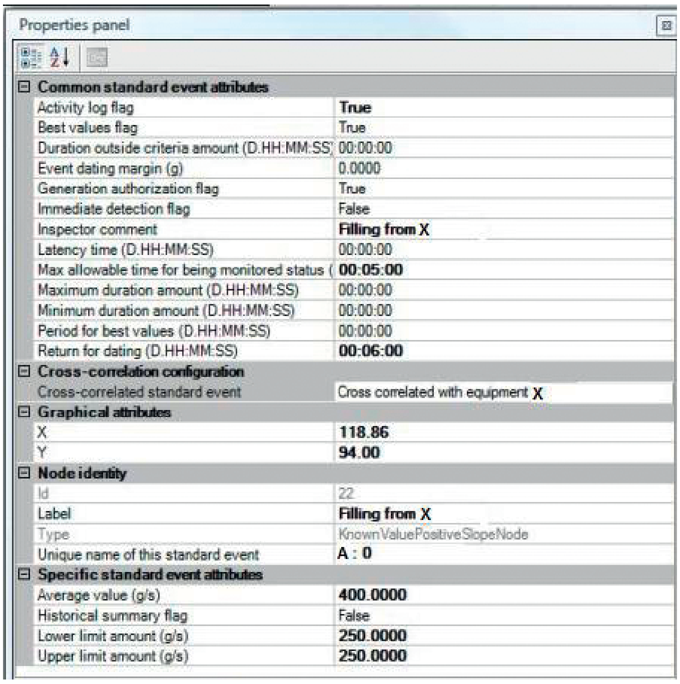
Configuration and parameter setting (parametrisation) have been envisaged for approximately 45 vessels, excluding the extractors for which overall pattern for the 1st and 2nd extraction cycles is foreseen to be monitored based on the signal from the neutron detectors. In addressing the configuration and parameterization work, 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.

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.

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 impor-



Properties panel

Common standard event attributes	
Activity log flag	**True**
Best values flag	True
Duration outside criteria amount (D.HH:MM:SS)	00:00:00
Event dating margin (g)	0.0000
Generation authorization flag	True
Immediate detection flag	False
Inspector comment	**Filling from X**
Latency time (D.HH:MM:SS)	00:00:00
Max allowable time for being monitored status (	**00:05:00**
Maximum duration amount (D.HH:MM:SS)	00:00:00
Minimum duration amount (D.HH:MM:SS)	00:00:00
Period for best values (D.HH:MM:SS)	00:00:00
Return for dating (D.HH:MM:SS)	**00:06:00**
Cross-correlation configuration	
Cross-correlated standard event	Cross correlated with equipment X
Graphical attributes	
X	**118.86**
Y	**94.00**
Node identity	
Id	22
Label	**Filling from X**
Type	KnownValuePositiveSlopeNode
Unique name of this standard event	**A : 0**
Specific standard event attributes	
Average value (g/s)	**400.0000**
Historical summary flag	False
Lower limit amount (g/s)	**250.0000**
Upper limit amount (g/s)	**250.0000**

Figure 7: Properties window of the standard event parameters

tant 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”.

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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Uncertainty estimation in nuclear material weighing

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Abstract

The assessment of nuclear material quantities located in nuclear plants requires knowledge of additions and subtractions of amounts of different types of materials. Most generally, the quantity of nuclear material held is deduced from 3 parameters: a mass (or a volume of product); a concentration of nuclear material in the product considered; and an isotopic composition.

Global uncertainties associated with nuclear material quantities depend upon the confidence level of results obtained in the measurement of every different parameter. Uncertainties are generally estimated by considering five influencing parameters (ISHIKAWA's rule): the material itself; the measurement system; the applied method; the environmental conditions; and the operator.

A good practice guide, to be used to deal with weighing errors and problems encountered, is presented in the paper.

Keywords: weighing, nuclear material, uncertainty, error, non destructive measurement

1. Introduction

Domestic and international regulations regarding nuclear materials impose rules requiring each operator to know continuously the location, quality and quantity of these materials. Physical follow-up is based in particular on measurements and analysis, carried out at key measurement points of the processes implemented in the plant and impacting these materials. Three types of measurements are generally performed:

- a weighing, to determine a quantity of product (for example: UF_6, uranium nitrate, uranium and/or plutonium oxides …);
- a concentration determination, to know the quantity of nuclear material (U, Pu) contained in the product; and
- an isotopic measurement, to determine ^{235}U in the case of uranium compounds.

The required performance of the measurement system (method, technical means…) depends on the final objective to be reached. These three quantities should be well-characterised (accuracy, uncertainty…). The quality of their knowledge and determination depends essentially on:

- the sampling representativeness;
- the measurement method used; and
- the performance of the equipment employed.

This paper defines some factors to be taken into account when using weighing machines. It focuses on different points that the operator has to consider in order to define and optimise the measurement system. The first step for the operator must be to define the measuring problem, in particular:

- the measurand, i.e. its definition and the expression and the unit of the result arising from measurement (for example a mass of product expressed in kg) and the types of products which will be measured (powder, solids, liquid…);
- the technical specifications required (maximum permissible measurement errors : MPE, uncertainties…) and the normative and contextual constraints.

This reflection determines the choice of the measurement system and the resources to be used in setting it up.

2. Definitions [1], [2], [3]

Mass – real mass: the mass (or real mass) m of an object is a physical, constant and intrinsic data of this object; it is equal to the product of its density r_m by its volume V ($m=r_mV$).

Apparent weight – apparent mass: weighings are generally carried out in the atmosphere (air of density ρ_a). The balance measures a force W called the apparent weight. This apparent weight is the sum of two opposed forces, a weight (W=mg) and a force F due to Archimedes's buoyancy ($F=\rho_aVg$).

Conventional mass: the conventional mass m_c of a body (of (real) mass m and density ρ_m) is the mass of a fictitious standard of density ρ_0=8000 kg/m^3 that balances this body under conditions conventionally chosen: an air of density ρ_{a0}=1,2 kg/m^3 and a reference temperature t=20 °C.

Body	ρ_m: density (kg/m3)	V: volume (cm³)	$\rho_a V$: air buoyancy correction/ gravity (g)	m_{app}: apparent mass (g)	m_c: conventional mass (g)	m: (real) mass (g)
Platinum	21500	47	0,056	999,944	1000,094	1000,000
U metal	19000	53	0,063	999,937	1000,087	1000,000
UO_2 sintered pellets	10500	95	0,114	999,886	1000,036	1000,000
Stainless steel	8400	119	0,143	999,857	1000,007	1000,000
Reference	**8000**	**125**	**0,150**	**999,850**	**1000,000**	**1000,000**
Aluminium	2700	370	0,444	999,556	999,706	1000,000
Solid UF_6 (t= 20°C)	5100	196	0,235	999,765	999,915	1000,000
$U0_2$ powder	2000	500	0,600	999,400	999,550	1000,000
Uranium nitrate solution	1400	714	0,857	999,143	999,293	1000,000

Table 1: Calculation of the conventional and apparent masses corresponding to the same (real) mass of 1000 g for various bodies

Error of indication: indication of an instrument minus the (conventional) true value of the corresponding mass. This error characterises the accuracy of a weighing instrument.

Accuracy class of a standard weight: standard weights (or sets of weights) are defined according to certain metrological requirements intended to maintain the uncertainties of mass values within specified limits. Nine weight classes are defined by the International Organisation of Legal Metrology (OIML): E1, E2, F1, F2, M1, M1-2, M2, M2-3 and M3. Weights of class E1 are the most accurate.

Accuracy class of a balance: balances are defined according to certain metrological requirements intended to maintain the error within specified limits. Four balance classes are defined (class I, II, III and IV) by the OIML. Balances of class I are the most accurate.

Actual scale interval d: value expressed in mass units giving the difference, between two consecutive reference marks for an analogical indication, or between two consecutive indications for a numerical indication. For a numerical instrument d is the quantification step of this instrument.

Verification scale interval e: value expressed in mass units and used for verification and classification of an instrument according to the legal metrology rules. Its value depends on the balance characteristics.

Maximum permissible measurement error (MPE): maximum difference, positive or negative, allowed by regulation between the indication of an instrument and the corresponding true value, as determined by reference standard masses or standard weights.

Error of eccentricity: given by the different measurement results for different positions of the same load on the balance weighing surface.

Measurement precision: closeness of agreement between indications or measured quantity values obtained by replicate measurements on the same or similar objects under specified conditions.

Accuracy: closeness of agreement between a measured quantity value and a true quantity value of a measurand.

3. Weighing of a body – Expression of the result

Given a scale previously adjusted with known conditions, I_o is the indication of weighing before deposit of the body and I_{load} is the indication after. The relationship $DI = I_{load} - I_o$ represents the net weighing result.

3.1. Real mass

If the balance has errors of indication E_I which were determined by using standards in conformity with recommendation R111 of the OIML, if the scale is adjusted just before using it, and if the error E_I was corrected before, the (real) mass m of the weighed body is given by:

$$m \approx (\Delta I - E_I)\left[1 + \rho_a\left(\frac{1}{\rho_m} - \frac{1}{\rho_0}\right)\right] \approx \Delta I - E_I + \Delta I \cdot \rho_a\left(\frac{1}{\rho_m} - \frac{1}{\rho_0}\right) \approx \Delta I + \Delta I.\rho_a.\left(\frac{1}{\rho_m} - \frac{1}{\rho_0}\right)$$

3.2. Conventional mass

Using the same notation and assumptions as previously, the conventional mass m_c of the weighed body is given by the relationship:

$$m_c \approx (\Delta I - E_I)\left[1 + (\rho_a - \rho_{a0})\left(\frac{1}{\rho_m} - \frac{1}{\rho_0}\right)\right] \approx \Delta I - E_I +$$

$$\Delta I \cdot (\rho_a - \rho_{a0})\left(\frac{1}{\rho_m} - \frac{1}{\rho_0}\right) \approx \Delta I + \Delta I.(\rho_a - \rho_{a0}).\left(\frac{1}{\rho_m} - \frac{1}{\rho_0}\right)$$

4. Influencing factors

The operator must always keep in mind the required objective of the measurement process. Some criteria (performance, uncertainties…) must have been proposed as indicators to demonstrate that:

- the process complies;
- the necessary controls are determined; and
- the corresponding means of measurement (method, equipment) are defined.

Different factors influence directly the quality of measurements and the associated uncertainties:

- the environmental conditions;
- the standard weights used;
- the operator's competence;
- the instruments used; and
- the weighing methods implemented (simple or double weighing).

The cause-effect diagram (fig.1), called the Ishikawa's diagram (or 5 M diagram), gathers the principal quantities to consider. Some variables may, of course, appear negligible depending on the measurement problem definition; they must however be identified and indicated.

4.1. Measurand

The measurand definition and its characteristics have direct influence on the weighing result and the associated uncertainty. The measurand must be defined carefully, with its unit specified and the most complete possible list of influencing elements.

4.1.1. Definition

According to the desired defined measurand (conventional mass or (real) mass), the air buoyancy correction is different and the uncertainty calculation must take it into account.

4.1.2. Temperature

A variation of the body temperature causes variations of volume and thus variations of apparent mass. It is necessary to know the cubic dilation coefficient α (expressed in $°C^{-1}$) of the weighed body, to evaluate the real volume at temperature t: $V_t = V_{20}(1+a(t-t_0))$ with $t_0 = 20°C$.

4.1.3. Body density

The product to be weighed can be composed of different bodies having various properties, in particular densities. It is advisable to calculate an equivalent density r_{eq} for the whole body (composed of the container, nuclear material,

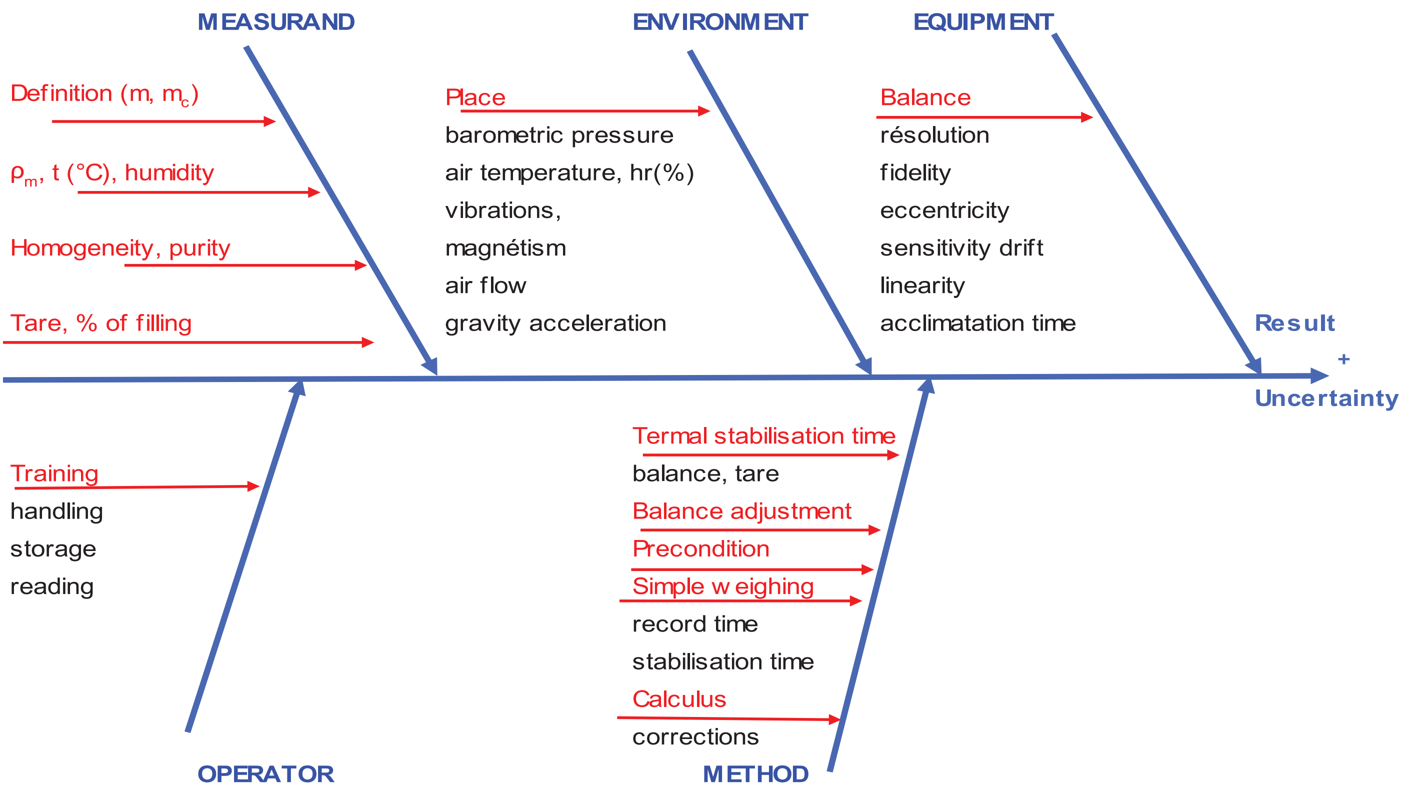


Figure 1: Ishikawa's diagram (or 5 M diagram)

air…) to apply correctly the buoyancy correction. For example, a steel container containing material X and air can be seen as a homogeneous body of density r_{eq} , mass m_{total} and volume V_{total} with the following expression:

$$\frac{1}{\rho_{eq}} = \frac{1}{m_{total}} \left(\frac{m_{steel}}{\rho_{steel}} + \frac{m_{air}}{\rho_{air}} + \frac{m_X}{\rho_X} \right), \; m_{total} = m_{steel} + m_{air} + m_X$$

and $V_{total} = V_{steel} + V_{air} + V_X$

The graph below shows the importance of the densities of bodies in weighing. In nuclear material safeguards, three domains are particularly distinguished:

- densities extending from 1400 to 5000 kg/m³; this corresponds to material in liquid form or powders;
- densities close to 11000 kg/m³; this essentially corresponds to sintered pellets; and
- densities about 19000 kg/m³ corresponding to U in metal form.

So, for example, in a UO_2 fuel fabrication plant, we suppose that uranium in powder form is the input material. This uranium is transformed into sintered pellets to produce the output materials, the fuels assemblies. A relative difference of (real) mass about $3{,}7.10^{-5}$ exists between both material quantities and has to be taken into account to estimate the real quantity of material having passed through the plant. This difference is only due to the difference in density between both products. This quantity seems to be very small, but is not negligible when taking into account the total quantities transformed.

4.1.4. Tare problem

If the container tare contributes to the final calculation, the corresponding measurand has to be specified. An empty container must be well characterised to avoid any problem due to the mass of possible product remaining inside the container (gas for example). For instance, two UF_6 cylinders of internal volume of 1 m³, one filled with air at atmospheric pressure and the other really empty (because of pumping), weighed in the same environment (balance, environmental conditions), present a difference in readings of about 1.2 kg.

4.2. Operator

The operator's competence can influence the measurement result. Several causes are at the origin of human errors, in particular, not complying with procedures (for example the acclimatisation durations to obtain thermal stability…), miscalculations, errors in manual records of results…

4.3. Environment

4.3.1. Levelling

A weighing device must always be level (the air bubble must stay in the mark centre). The bubble position must be

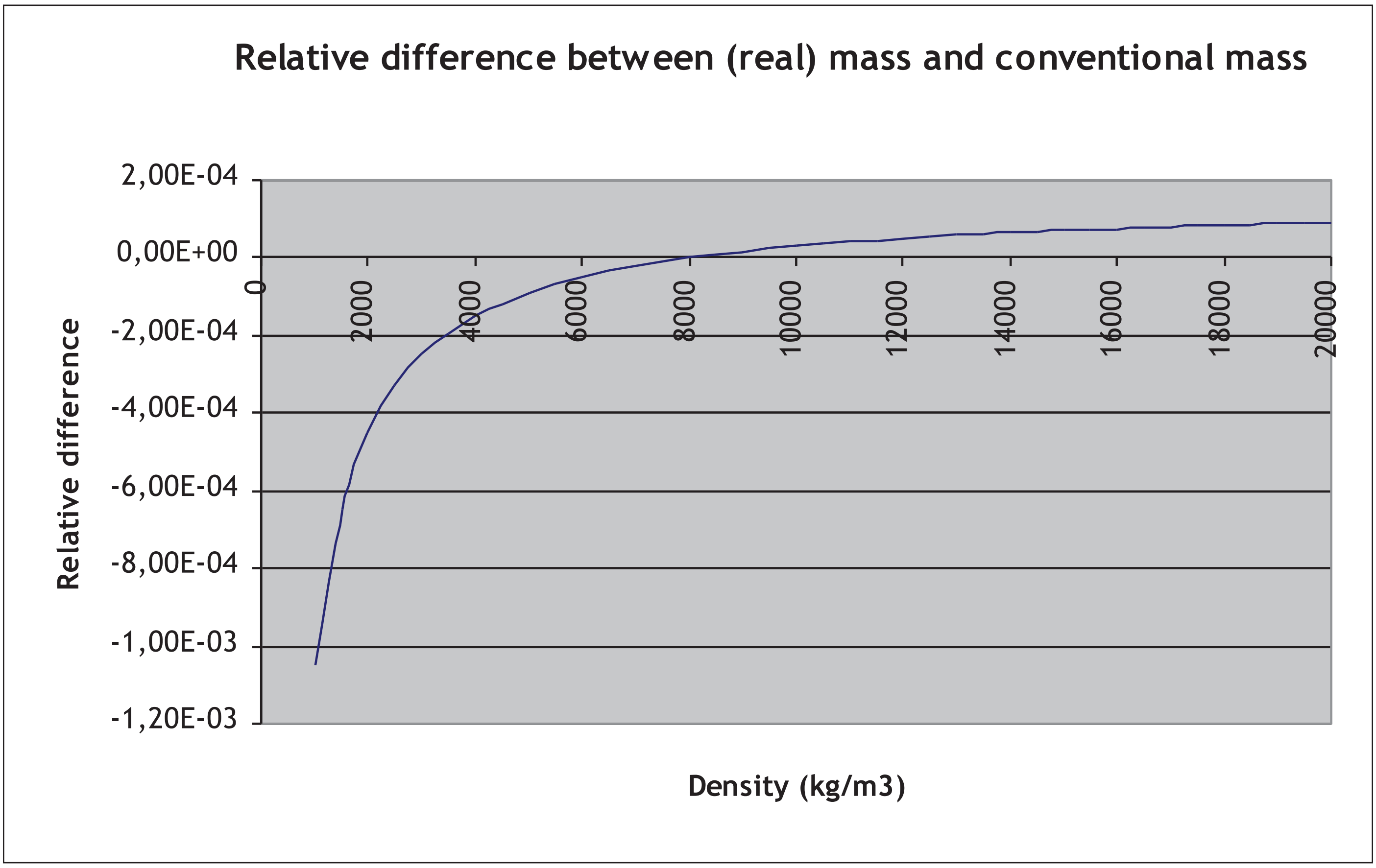
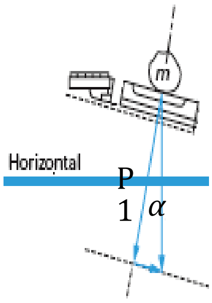


Figure 2: Relative difference between a mass and its conventional mass

corrected, if necessary. In this example, the balance measures only the P component, perpendicular to the pan.



4.3.2. Setting of the weighing device

While installing the weighing machine, attention should be paid to ensure that:

- the influence of drafts is limited (by moving the device away from doors, ventilation, heating sources and air-conditioning);
- the influence of direct radiation is controlled (by moving the balance far from windows);
- the weighing device is placed on a stable and rigid base (no vibration) and is protected from shocks during handling operations; and
- the weighing device is protected from static electricity and magnetism problems.

4.3.3. Environmental conditions

In all cases, the weighing device environment (air temperature, barometric pressure, humidity) must be stable. Indeed these parameters directly influence the balance sensor and the air density. The following simplified formula may be used to determine the air density ρ_a (kg/m³) according to the barometric pressure p (in hPa), temperature t (in °C) and relative humidity of the air h_r (in %):

$$\rho_a = \frac{0{,}34848 \cdot p - (0{,}009 \cdot h_r.\exp(0{,}061.t))}{273{,}15 + t}$$

This equation has a relative error of 2.10^{-4} for measurements in the range:
900 hPa < p < 1100 hPa, 10 °C < t < 30 °C and h_r < 80 %.

4.3.4. Local gravity acceleration

Electronic weighing devices measure the force induced by the body weighed but do not determine its mass. The gravity acceleration g depends on location (altitude and latitude). The balance indications thus depend on local g. By carrying out a scale adjustment (internal or external) on site, the operator can regulate correctly and automatically the influence of the local gravity. The formula hereafter may be used to determine the local g (in m/s²) according to the latitude *F* (in degrees) and altitude *H* (in metres) of the site.

$$g = 9{,}780318.(1 + 0{,}0053024\sin^2\Phi - 0{,}0000058\sin^2 2\Phi) - 0{,}000003085.H$$

For example, we consider a weighing device adjusted, calibrated and used in Fontenay-aux-Roses (near Paris). It indicates 10 kg for a mass of 10 kg. If this device is transported to Pierrelatte (in the south of France) but is not readjusted on this site, although the same mass of 10 kg is measured this mass does not generate the same force in both places mentioned due to the gravity g variations.

We have:

$$\frac{P_{FAR}}{g_{FAR}} = \frac{P_{Pier}}{g_{Pier}} \text{ so } P_{Pier} = P_{FAR}\frac{g_{Pier}}{g_{FAR}} = 10000 \times \frac{9{,}8052}{9{,}8084} = 9996.7g$$

Fontenay-aux-Roses	Pierrelatte
Latitude Φ=48°, altitude H=160 m	Latitude Φ=44°, altitude H=53 m
gFAR=9,8084 m.s-2	gPier=9,8052 m.s-2
PFAR=m.gFAR	PPier=m.gPier
Indication of the balance adjusted in FAR: 10000,0 g	Indication of the balance not re-adjusted on site: 9996,7 g

Table 2: Example of local gravity influence

4.4. Equipment

4.4.1. Weighing device

The balance performance affects the uncertainty of the final result. The principal factors to be considered are:

- the resolution of the scale;
- its sensitivity, its precision;
- its eccentricity limits; and
- its linearity.

The metrological characteristics of the balance must be considered in the evaluation of the instrument uncertainties. Four accuracy classes of balances (class I, II, III and IV) are defined according to their performance. Class I corresponds to a high accuracy apparatus used in a laboratory, class IV corresponds to current appliances. In the nuclear field, an operator generally needs a balance of class II for his physical follow-up of nuclear material. The devices of class I are used by laboratories carrying out very accurate analysis.

4.4.2. Choice of standards

To calibrate and verify balances, it is necessary to use either internal or external standards traceable to international standards. The use of magnetised standards of weights whose thermal equilibrium is not reached is a source of errors which must be avoided. The MPE of the standards used must be lower than 1/3 of the balance MPE.

Steps	Operation	Results obtained	Commentaries
1rst step	Determination of the indication error. Associated uncertainty	Indication error: E_I Expanded (k=2) uncertainty $U(E_I)$	Operation called calibration
2nd step	Determination of the balance uncertainty	Expanded (k=2) uncertainty U(IP) of the balance	Exploitation of calibration

4.4.3. Equipment transport

Any transport presents risks for the equipment. Care should therefore be taken that the transported balance is considered operational only if adequate controls (adjustment, verification and calibration) are carried out before starting measurements.

4.5. Measurement method used

In any method used for calibration and weighing, the number of measurements has a direct influence on the weighing result and its associated uncertainty. A Gauss weighing (double weighing) is more accurate than a simple weighing but requires two successive operations and more effort in uncertainty calculation.

The closer the calibration and verification conditions are to the real conditions of use, the less the results have to be corrected and the less important the uncertainty associated with the correction is.

This document only deals with simple weighing. Two procedures for using balances may be envisaged.

The first method consists of carrying out a single calibration in Fontenay-aux-Roses and not re-calibrating the balance after each change of location. It is then necessary to correct systematically the indication of the balance (effect of gravity, temperature and pressure of the ambient air, etc.), which leads to additional measurements in order to be able to make the right corrections. This method is not applied by the inspectors.

The second method, used for inspections because it is simpler, entails a re-calibration[1] of the balance on each site after each change of location, so as to adapt to the local conditions. Use of the initial calibration curve providing the uncertainty of the weighing instrument established previously at the Fontenay aux Roses laboratory is not theoretically possible since the parameters of the balance are modified at each re-calibration. Strictly speaking, it would be necessary to carry out a calibration and recalculate the corresponding uncertainty. In practice, the standard uncertainty (of the form $u = MPE_{bal}/\sqrt{3}$) linked to the operations of verifying the compliance of the balance carried out after each re-calibration is predominant and is added to the uncertainty component linked to the initial calibration of the form $u=a+b^*\Delta I$, which is conserved.

The calibration operation involves carrying out an internal or external re-calibration. Following this calibration, the operator carries out a repeatability, eccentricity and linearity test.

5. Determination of the uncertainty linked to the weighing instrument

5.1. Determination of the uncertainty linked to calibration

The laboratory uses the COFRAC (French Accreditation Committee) approach to determine the uncertainty linked to weighing. This method is accredited in accordance with the ISO 17025 Standard.

The uncertainty associated with a weighing result is a function of different operations: calibration with standards and conditions of use. Two successive steps are distinguished to calculate the measurement uncertainty.

- The 1st step is used to determine the error of indication E_I (internal calibration of the balance and the associated expanded (k=2) uncertainty $U(E_I)$.
- The 2nd step is used to determine the expanded uncertainty U(IP) of the measurement.

The calculation takes into account:

- the reading resolution of the balance;
- the weighing repeatability;
- the uncertainties of standards;
- the effects of eccentric loadings; and
- the temperature and density of the air.

The error of indication E_I is given for a load. It is obtained by difference between the result of the simple weighing of a standard load and its certified value. The indication error E_I is given with one or more load values belonging to the domain of use. Loads may be applied with growing and/or decreasing values.

[1] Following an internal or external adjustment of the device on the site, the operator carries out, with standards, a repeatability test (4 to 5 measurements), an eccentricity measurement and a linearity test (on the balance range). The resolution with and without a load intervenes also. The acceptance criteria for these tests are that each difference observed between any result read and the corresponding load used is lower than the balance MPE (maximum permissive error).

The temperature t also directly influences the sensor response according to the load deposited on the balance pan. The relative uncertainty due to the temperature effect on the weighing instrument is given by the relation: $u(t)_{rel}=C.\Delta t/\sqrt{3}$ (uniform distribution) where C is the variation coefficient of the instrument slope versus the temperature and Δt the variation in temperature during calibration.

This coefficient C depends on the type of instrument: it is often given by the manufacturer. In case of the absence of data, table 2 (below) gives acceptable values.

Maximum number of steps e of the instrument m=Load/e	Maximum variation coefficient versus temperature for approved instruments (°C-1)	Maximum variation coefficient versus temperature for other instruments (°C-1)
1000	250 x 10-6	2 500 x 10-6
10000	25 x 10-6	250 x 10-6
100000	5 x 10-6	50 x 10-6
>100000	1,5 x 10-6	15 x 10-6

Table 3: Variation coefficient of the balance sensitivity versus the temperature

5.2 Determination of the uncertainty linked to verification

After having correctly installed the equipment in a suitable place and waited for sufficient time to allow the balance and the calibration weights to reach ambient temperature, the inspector carries out a re-calibration and performs a verification[2] that it is working correctly by weighing said calibration weight. The inspector completes the specific form for the balance used, following the instructions given (see table 4). The result of these controls entails a finding: the balance is compliant or not.

5.3 Determination of the total uncertainty linked to the instrument

It suffices to sum quadratically the different uncertainties obtained in § 5.1 and § 5.2.

One obtains a relation of the form:
U_{total} (k=2) (g) = $[(a + b^*\Delta I)^2+(2^*MPE_{bal}/\sqrt{3})^2$

Since the uncertainty linked to the verification of balances evolves in the form of a "tunnel", the observed discontinuities need to be eliminated by linearising piece by piece the uncertainty over the whole range of use.

5.3.1 Application to the balance LE34001P

The range of use of the balance LE34001P, of class II (verification level e=1 g), multi range (3 different reading levels d

[2] The criterion for verifying the correct operation of the balance stipulates that, for each individual measurement carried out, the deviation observed between the value read and the corresponding load placed on the tray of the balance lies within the interval ± 1 MPE_{bal} for the load in question. This consequently leads to a possible error of ± 1 MPE_{bal}. Assuming that this observed deviation follows a rectangular probability law with zero mean, the associated standard uncertainty is close to $MPE_{bal}/\sqrt{3}$. This uncertainty varies as a function of the range of use and, given the variation of MPE, changes according to the approximate form of a "tunnel", with discontinuities.

1 - External-interal Calibration

Number of weighings	Load R real or simulated (g)	MPE (g)	Indication L read on the balance (g)	Difference E = R-L (g)
Calibration	10.000	2	10.000,0	0,0

2 - REAPEATABILITY

Number of weighings	Load R real (g)	MPE (g)	Indication L read on the balance (g)	Difference E = R-L (g)
1	10.000	2	10.000,2	-0,2
2	1.000	2	10.000,0	0,0
3	10.000	2	10.000,2	-0,2
4	10.000	2	10.000,2	-0,2
5	10.000	2	10.000,2	-0,2

3 - ECCENTRICITY

Location	Load R real (g)	MPE (g)	Indication L read on the balance (g)	Difference E = R-L (g)
1	10.000	2	10.001,2	-1,2
2	10.000	2	10.000,2	-0,2
3	10.000	2	10.000,6	-0,6
4	10.000	2	10.001,2	-1,2

4 - ACCURACY

Load chosen	Load R real (g)	MPE (g)	Indication L read on the balance (g)	Difference E = R-L (g)
Min	3.000	1	3000,1	-0,1
0,25*Max	10.000	2	10000,2	-0,2
0,50*Max	20.000	2	20000,5	-0,5
0,75*Max	30.000	3	30001,5	-1,5
Max	34.000	3		

Conformity : Yes

Table 4: Example of records from the verification of the balance LE34001P

as a function of the load: 0,1, 0,2 and 0,5 g), lies between 50 and 34000 g.

The expanded uncertainty (k=2), expressed in g, associated with the calibration of the weighing instrument in Fontenay-aux-Roses, is estimated by the formula:

U_{bal}=0,08+0,0000331*R where R is the uncorrected result (in g) given by the balance.

In accordance with the method described in § 5.2, the expanded uncertainty linked to the verification is close to 2*MPE_{bal}/√3. Discontinuities appear at 5000 and 20000 g.

It suffices to sum quadratically the previous results to obtain the total uncertainty linked to the balance. One thus obtains:

$$U_{total}\ (k=2)\ (g) = [(0{,}08+0{,}0000331*R)^2+(2*MPE_{bal}/\sqrt{3})^2]^{0,5}$$

The following table gives the total uncertainty U_{total} (k=2) expressed in g, over the whole weighing range.

Range (g)	[50	5000]	]5000	20000]	]20000	34000]
d_0(g)	0,1	0,1	0,1	0,1	0,1	0,1
d(g)	0,1	0,1	0,1	0,5	0,5	0,5
MPE_{bal}(g)	1,0	1,0	2,0	2,0	3,0	3,0
COFRAC U_{bal} (0,08+0,0000331*R, k=2) (g)	0,1	0,2	0,2	0,7	0,7	1,2
U(verification, k=2)= 2*EMT_{bal}/√3 g)	1,2	1,2	2,3	2,3	3,5	3,5
$U_{total\ use}$ (k=2) (g)	1,2	1,2	2,3	2,4	3,5	3,7

The final equations used to determine the uncertainty $U_{total\ use}$ of any weighing result is given above:

Range (g)	[50 – 5000]	]5000 – 20000]	]20000 – 34000]
$U_{total\ use}$ (k=2) (g)	1,16+4,63.10-⁶*R	2,29+6,88.10-⁶*R	3,37+8,94.10-⁶*R

6. Application to measurements carried out during inspections

As has been seen in § 2, the weighing results may be expressed in real weight or in conventional weight. The operators and inspectors respectively use, for physical monitoring and accounting for nuclear materials in their installations and controls on site, the conventional weight[3] of bodies directly measurable with a calibrated balance.

Finally, the uncertainty to be considered for any unknown quantity weighed must take into account the uncertainty linked to the instrument $U_{total\ use}$ (see above) and the uncertainty associated with the weighed material itself (the measurand). This must be quantified on a case by case basis, taking into account the factors discussed in chapters 3 and 4. The final uncertainty is the quadratic sum of each term. This uncertainty is given with an expansion factor of 2.

In practice, the measurand is often poorly known and poorly defined and the characteristics of the weighed body are imprecise. It may thus be very difficult to estimate, for example, the weight of nuclear material contained in the weighed body. Each case is a specific case and it is necessary to carry out a specific calculation to determine an element as a function of the different components of the measurand. The associated uncertainty calculation is thus specific and must be treated on a case by case basis.

For example, in a cylinder of UF_6, only the net weight of uranium contained in the cylinder needs to be known precisely with regard to regulations. In general, this weight is obtained from the difference between the weight of the full container and the corresponding tare weight. The gross weight is measured directly with a calibrated balance, whereas the tare is determined from a traceability system and, in particular, the indications etched directly on the cylinder. However, no details are provided regarding the conditions under which this tare has been determined (cf. § 4.1.4).

Experience shows that in this case the uncertainty due to the weighing instrument is negligible vis-à-vis the uncertainties linked to the lack of knowledge of the measurand.

7. Conclusions

Weighing is often considered a priori as a very simple operation, but it is not. This depends on the required objective (performances, needs...). In carrying out a 5 M standard analysis (Ishikawa's diagram) in order to evaluate the different uncertainty sources of the process, the operator realises that many factors are to be taken into account. Three factors have to be considered in priority:

- the intrinsic performances of the instrument;
- the environmental conditions of calibration and current use; and
- the knowledge of the weighed products.

The uncertainties obtained must be specially treated and calculated in each case.

References

[1] OIML R76- 1 Edition 2006. Non-automatic weighing instruments Part 1: Metrological and technical requirements – Tests

[2] VIM international vocabulary of metrology

[3] OIML R111-1 Edition 2004, Weights of classes E1, E2, F1, F2, M1, M1-2, M2, M2-3 and M3 Part 1: Metrological and technical requirements.

[3] Note: Unlike inspection controls, which are only monitoring controls, it is necessary to carry out calculations of Material Uncounted For (MUF) and associated uncertainties from real weights. The use of conventional weights generates biases in the case where the nuclear materials undergo physical-chemical transformations and thus variations in density.

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.

The applicability of SAR-data for Safeguarding purposes is well described in existing publications, e.g. by Loreaux [1], but for the first time, this study can be based on High-Resolution SpotLight stacks of 10 and more scenes.

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. ESA's C-Band satellites and ALOS-PALSAR can not provide recent data, however they provide an extremely valuable archive and new sensor will be launched in the years to come. 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

TerraSAR-X Overview	
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 [2].

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) [3].

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.

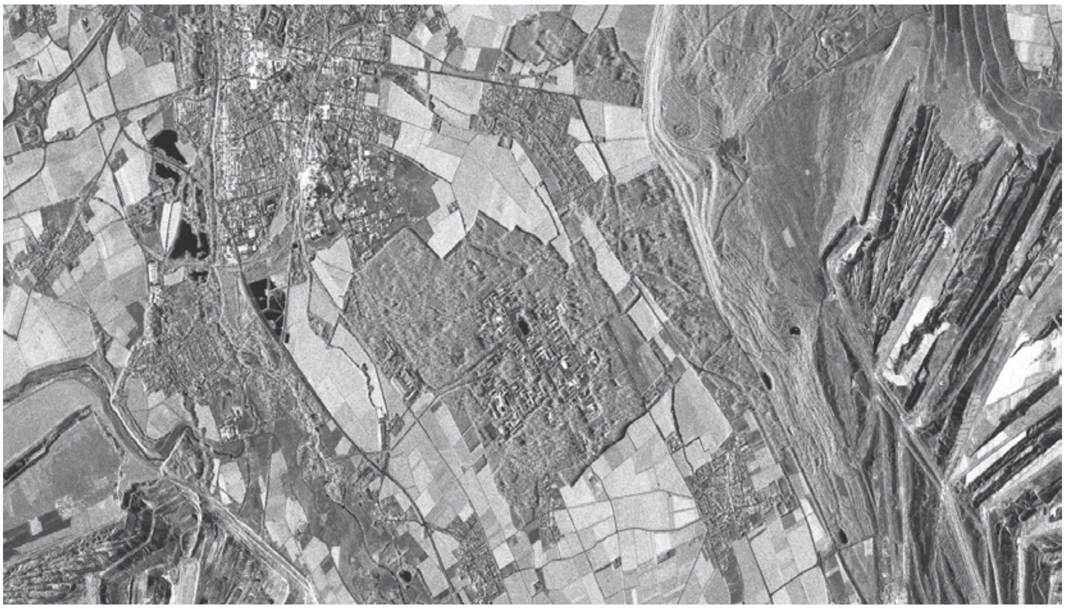

Figure 1: Example of the amplitude information of a single High-Resolution Spotlight TerraSAR-X scene, presented in range (horizontal) and azimuth (vertical) coordinates.

3.1. Exploiting amplitude of acquisitions

The amplitude of one or more SAR acquisitions is mainly independent of atmospheric conditions, completely independent of lighting-conditions (day-night capability) and can already be used for safeguarding applications. However, several specific aspects of RADAR-acquisitions have to be considered.

- The backscatter of a certain feature is strongly dependent on the material it consist of, it's shape and a possible coverage by other materials, such as snow. Generally, man-made features, especially metallic and rectangular, provide a backscatter, that is much larger than the one of bare ground, vegetated areas and water bodies. As most of the Safeguarding interest lies with buildings and similar structures, SAR acquisitions provide an excellent source of information. Possible snow-cover affects the backscatter of a whole scene, but manly the ground, so buildings, especially with vertical walls, can still be identified.
- Complex buildings can cause multi-bounce effects, which are difficult to interpret. An inverse approach, namely using a building model and simulation the backscatter effects as described by Auer [4] can be applied to better understand these multi-bounces. Additionally, dual-polarisation acquisitions can be used to separate different types of reflections.
- Speckle noise can make image interpretation more difficult, but can easily overcome be creating a temporal mean images (this strongly affects the response time) or applying other well established filter algorithms.

A single radar acquisition offers, besides the "imaging" of the area or geodetic localisation of certain features as shown in Eineder [5], 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 [6].

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), or by ordering the acquisition as geocoded amplitude image and measuring the values in a suitable GIS software. However, this method is only applicable, if the complete vertical wall of the monitored building is visible and not covered by vegetation or other building in close vicinity.

In areas with a high building density, the side-looking geometry of a SAR-system can cause a near range building to cover the base of a far range building. In this case the above-mentioned method can not be used to retrieve building height information.

Using a second image from an opposite viewing geometry (ascending/descending-combination) can overcome this limitation by only increasing the response time for approx. two days in case of TerraSAR-X's 11 day-orbit cycle.

If two images of the same area are acquired from different directions, the geometric effects (layover, foreshortening

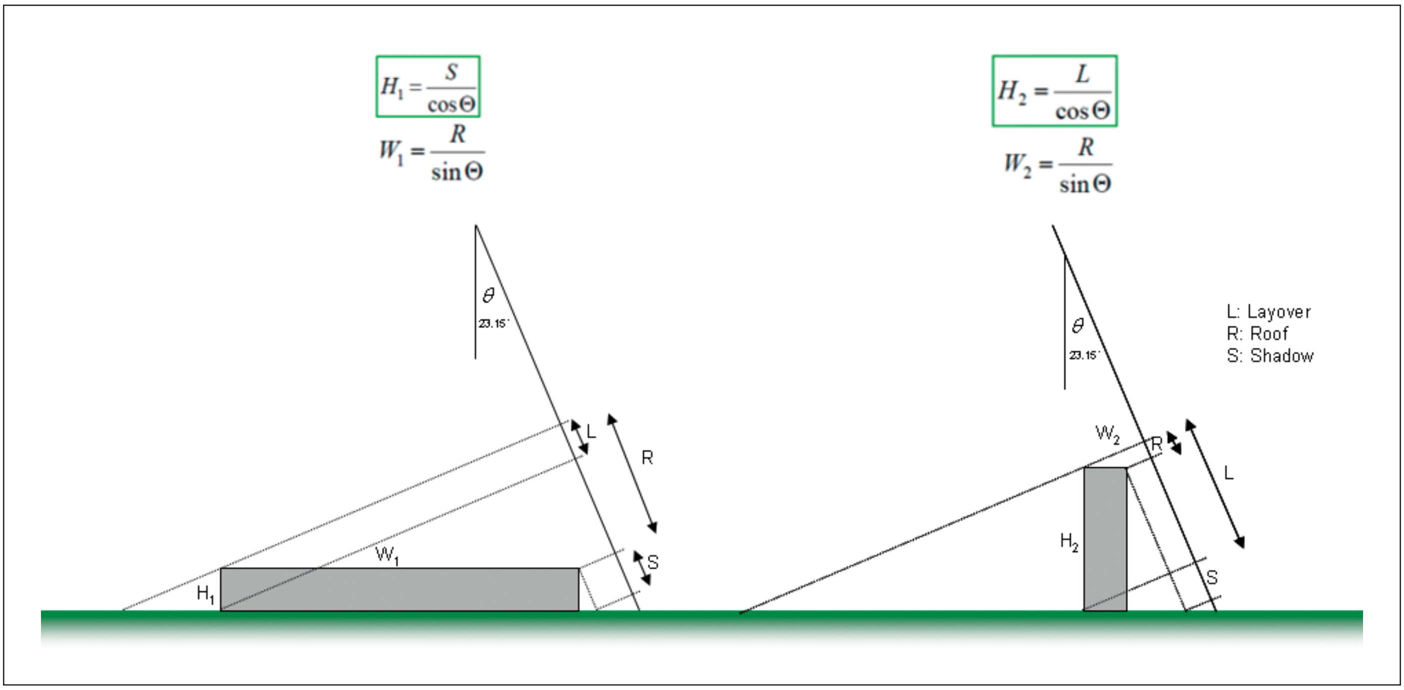

Figure 2: Geometric relation in SAR-imaging of buildings, showing the trigonometric relations of real height and width to SAR-image layover and shadow.

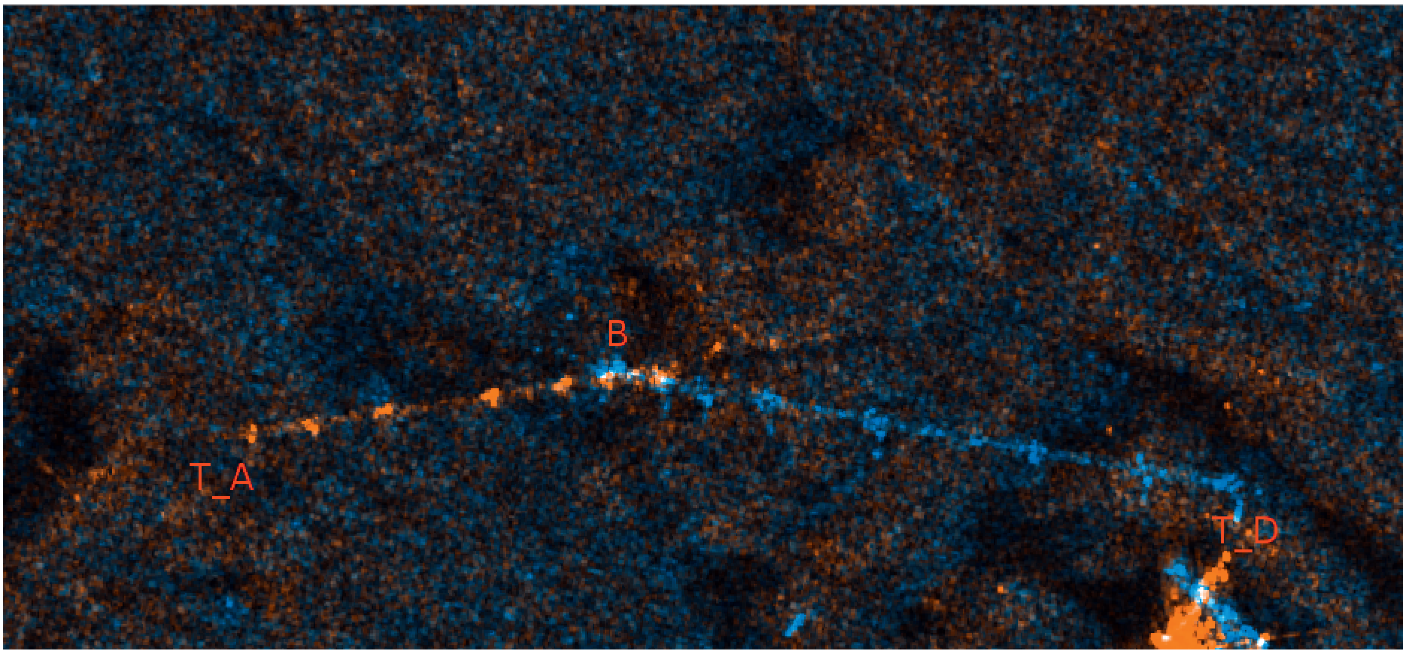

Figure 3: Amplitudes of an ascending (orange) and descending (blue) pass, geocoded and combined, showing the typical tilting towards the satellites LOS, B: Bottom. T_A Top ascending, T_D Top descending

and shadow) appear in different directions as well, so that e.g. roof structures are migrating away from each other as seen in Figure 3.

Both acquisitions have to be geocoded using the same DEM, as correction-terms are depending on the local height at the monitored feature, which is not always known with the necessary accuracy.

Figure 3 shows a mast used for weather-monitoring in an ascending/descending combination. The tilting towards the sensor is clearly visible, and the different amount of layover indicates different incidence angles. As the image is geocoded, the orbit inclination of TerraSAR-X is visible in the direction of the layover. In the example above, both methods to measure the height are applicable.

Measuring the length (strictly in LOS) of the layover area in an a descending, geocoded image with an incident angle of 22°, results in 302m. Using this distance and the incident angle (θ_1 = 22°) as an input for

$$h = \Delta x \cdot \tan\theta_1$$

the height calculation results in 122.3 m, which is in very good accordance with the real height of 124 m. This method is very robust, but both, the bottom and the top of the feature of interest have to be clearly visible in the image. If the bottom of a feature is not visible, which is probable in industrial complexes, a second acquisition acquired from a crossing orbit (Figure 3) is necessary.

Height estimation can then be performed by measuring the displacement of the top of the building (in x and y direction) in the different acquisitions using the heading (β_1 and β_2) and the incident angle (θ_1 and θ_2) of the used acquisitions by applying the following formula.

$$\sqrt{\Delta x^2 + \Delta y^2} = h\sqrt{\left[\left(-\frac{1}{\tan\theta_1}\cos\beta_1 + \frac{1}{\tan\theta_2}\cos\beta_2\right)^2 + \left(\frac{1}{\tan\theta_1}\sin\beta_1 + \frac{1}{\tan\theta_2}\sin\beta_2\right)^2\right]}$$

In the example above (Figure 3), shift in x direction results in 260.7 m and 23.9 m in y direction.

	Ascending	Descending
Incident Angle [°]	47.56	36.15
Heading Angle [°]	346.0	194.0

Table 1: Observation directions form asc/desc combination

Together with the observation angle from Table 1, the height of the mast can be calculated with 120.9 m, independently of the base visibility.

Accuracies of this method are depending on the resolution of the used acquisitions and the manual selection of the base- and top-points, which normally can be done with an error of very few pixels, but this implies the use of the highest resolution available (in case of TerraSAR-X: High Resolution SpotLight with a resolution of up to 1 meter).

3.2. Amplitude changes between two ore more 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, min-

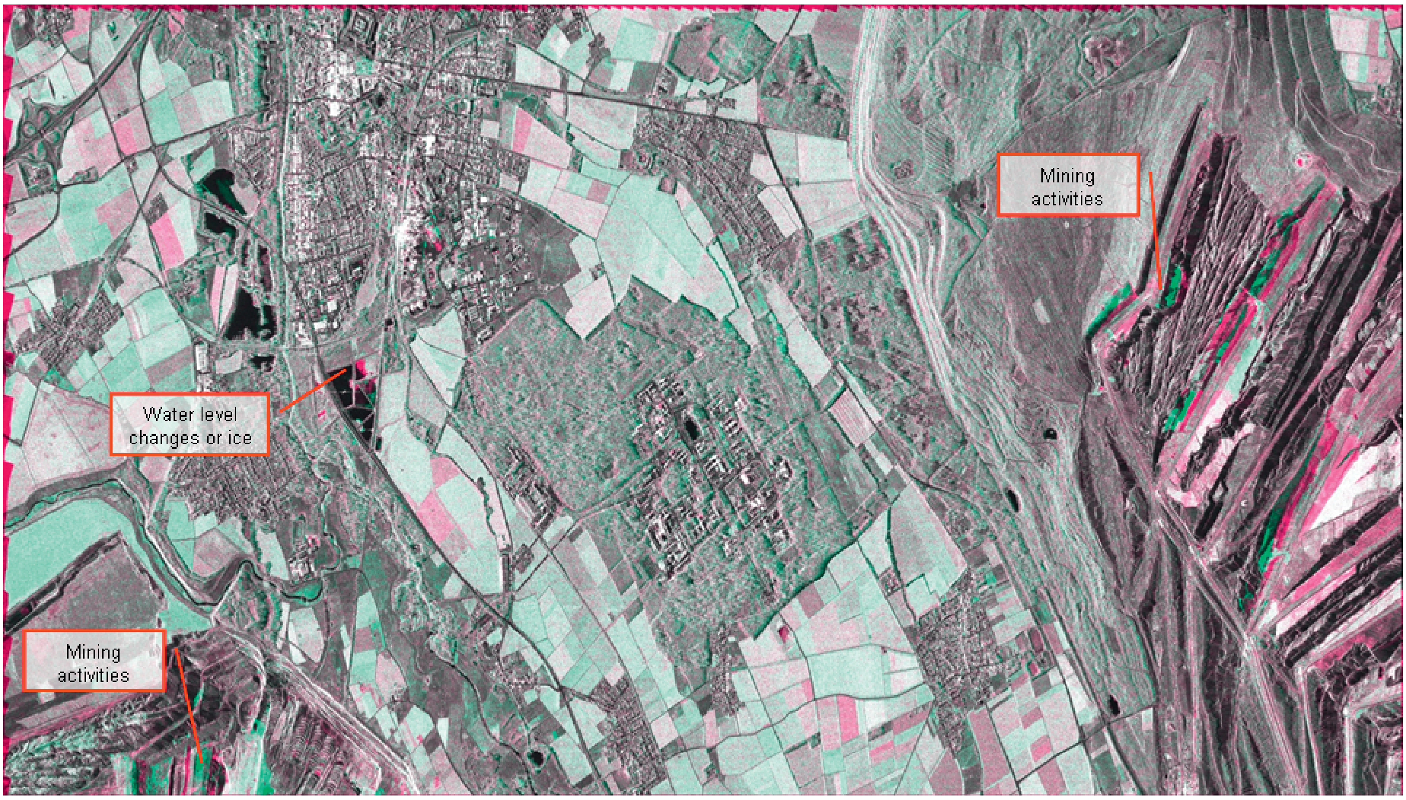

Figure 4: Amplitude changes between two SAR-Acquisitions (temporal distance 22 days), showing mining activities and water level changes, (red: brighter in 1st image, green: brighter in 2nd image).

ing activities and certain reflecting objects like cars, trains or containers can be identified (Figure 4).

Metallic objects, especially like containers, ships, cars, wagons, etc. show strong backscatter and are therefore easily identifiable in radar acquisitions. Figure 5 shows the relocation of containers, between the two acquisitions with a temporal separation of 33 days and mining activities at an opencast pit.

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 6).

Interferometric results are mainly limited by decorrelation, the amount is dependant on temporal and perpendicular baseline of the interferometric pair, the wavelength of the SAR and the features monitored in the scene. The coherence, a quality estimate for a interferograms, generally decreases with increasing baselines and decreasing wavelengths.

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 [8].

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 [9] can be performed (Figure 7).

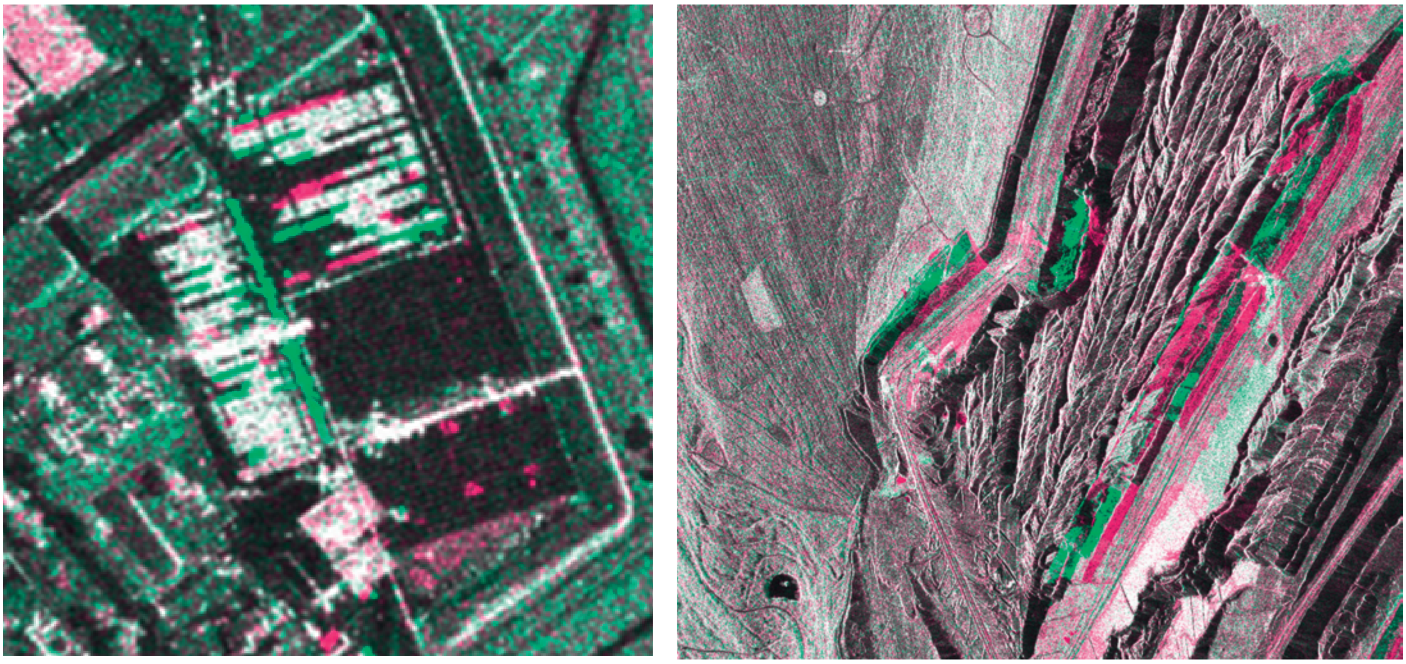

Figure 5: Amplitude changes showing relocation of containers (left) and mining activities (right), (colours like in Fig 3, temporal distance 33 days, respectively 22 days).

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.

PS-Processing is applicable mainly in environments dominated by man made feature, grassland and woods do not provide a sufficient number of stable targets. The number of PS can be artificially increased by distributing Corner-reflectors, which is not always possible. The accuracy and point density of the retrievable point-cloud is ranging between LIDAR-results (1m or better resolution) and future TanDEM-X surface models, which will have a resolution of approximately 12 meters.

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

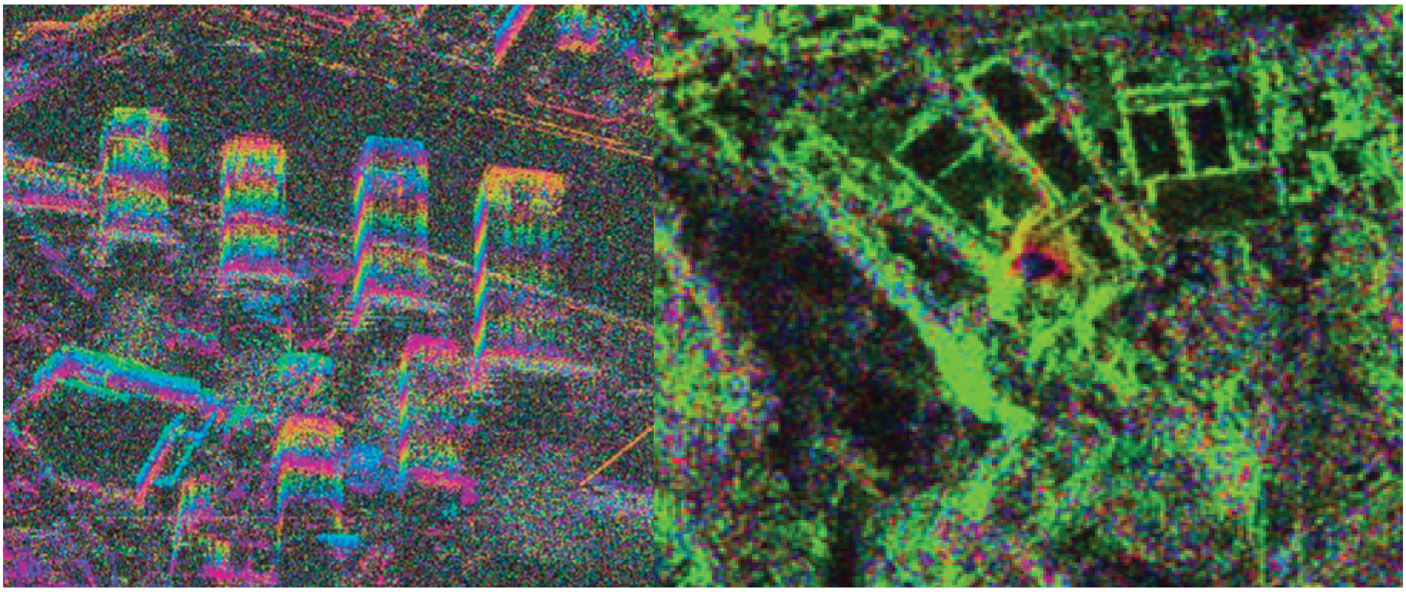

Figure 6: Left: Interferometric height measuring shown at skyscrapers in Tokyo, one colour-cycle corresponds to 43.8 m height, Right: Interferometric deformation measurement at Campi Flegrei, showing ~1 cm uplift over 11 days due to fumarolic activities, presented in Minet [7].

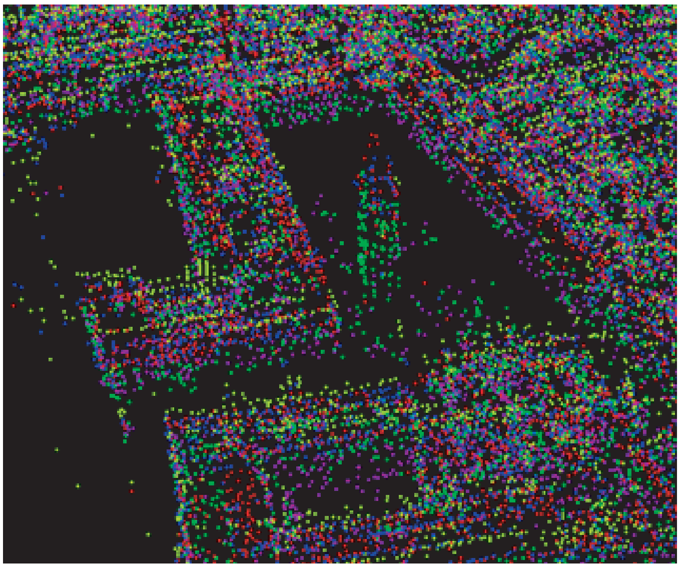

Figure 7: Combination of 5 PS-InSAR stack reconstructing the Campanile and Piazza San Marco in Venice, each colour representing one observation direction, processed by Hanisch [10].

waste rock and even the amount of the produced raw material can be estimated (Figure 8).

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.

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. DLR's TanDEM-X operationally acquires data since the beginning of 2011; however final DEM's are not available for now, since sever-

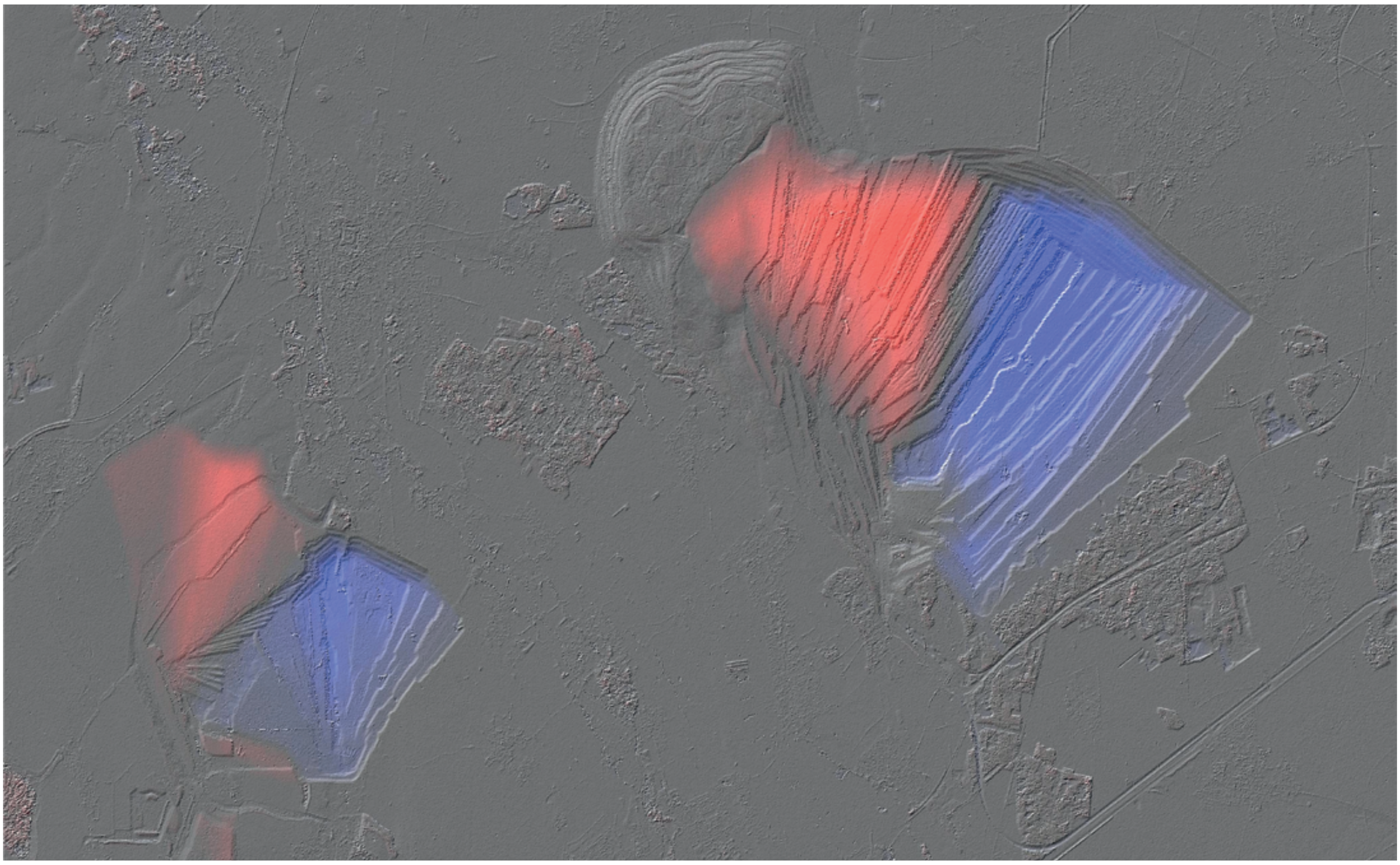

Figure 8: Mining Progress at Inden and Hambach opencast pits between 2000 (SRTM-Data) and 2010 (TanDEM-X data), red indicating waste rock deposition and blue indicating material extraction

al acquisitions with different imaging geometries and baselines have to be acquired. The defined resolution for TanDEM-X DEM products is 12 by 12 meters, may not be sufficient for high resolution building monitoring, but of course for monitoring mining activities. Integration and fusion of multi type data will be the main topic for the ongoing studies.

6. Acknowledgements

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Preparation and development of new Pu spike isotopic reference materials at IRMM

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Abstract

Reliable isotope measurements of nuclear material and the availability of reference materials with small uncertainties in the certified values are of great importance for safeguarding of nuclear materials. They provide the basis for a credible measurement system in the verification of states declarations of their 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, along with progress in modern analytical techniques. Some plutonium reference materials, however, have been on the market for decades and they need to be re-certified to smaller uncertainties. Moreover, new reference materials with appropriately small uncertainties in the certified values need to be made available enabling measurement laboratories to reduce their combined measurement uncertainties. Such high quality plutonium isotopic reference materials are essential for 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. An inter-calibration campaign has been set up at IRMM inter-linking selected plutonium spike IRMs. 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 IDMS. 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 re-certified 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. IRMM-049d was prepared from the same stock solution as its predecessor IRMM-049c, dating from 1996, but the new ^{242}Pu spike has certified values with smaller combined uncertainties. The traceability of the certified values to the SI is established through an unbroken chain of comparisons, all having stated uncertainties.

IRMM is also co-operating 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 system to safeguard nuclear activities in line with the Treaty on the Non-Proliferation of Nuclear Weapons (NPT) and the Euratom Treaty, including the respective implementing regulations, such as the Comprehensive Safeguards Agreements (INFCIRC 153, corrected) [1], the Additional Protocol (INFCIRC 540) [2] or the European Commission's Regulation on the application of Euratom Safeguards (No 302/2005).

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 accountancy. 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" [3]. The IAEA took over the concept of ITVs in the early 1990's from the ESARDA Working Group on 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. 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.

Some nuclear CRMs were approaching exhaustion; therefore a programme has been set up at IRMM to replace these materials. Two new nuclear materials, IRMM-046c and IRMM-049d, used in nuclear material analysis by IDMS, have been prepared. The certification of these new CRMs was part of an IRMM compatibility study inter-linking various plutonium spike CRMs on a metrological basis, applying state-of-the art measurement procedures [4]. Furthermore, IRMM prepares and certifies on a regular basis IRMM-1027 large-sized dried (LSD) spikes which are applied for the analysis of spent fuel solution at reprocessing plants.

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 the case of plutonium, the isotope pairs $^{241}Pu/^{241}Am$, $^{238}Pu/^{234}U$, $^{239}Pu/^{235}U$, $^{240}Pu/^{236}U$, and, possibly, 242Pu/238U can be used as "clocks". Some preliminary results for NBS SRM 946 will be presented here.

2. Need for new nuclear CRMs

The accurate verification of the plutonium and uranium amount contents in nuclear materials requires the continuous supply of well certified spike IRMs. The worldwide demands for certified spike materials have evolved considerably over the years, leading to the development of new plutonium and uranium reference materials with smaller uncertainties in the certified values.

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 from 1995. In addition, to guarantee future provision of these valuable spike materials to the nuclear measurement community, IRMM-049d, a highly enriched ^{242}Pu spike, has been prepared and certified by IDMS and another mixed uranium-plutonium spike, IRMM-046c, is currently in preparation. 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.

The isotopic reference material IRMM-1027 series have been used for the measurement of Pu and U amount content in dissolved fuel solution for some 20 years. They are designed for fissile material accountancy by Euratom Safeguards authorities at on-site laboratories at La Hague and Sellafield. A new set of IRMM-1027 LSD spikes, containing about 50 mg of uranium and about 1.8 mg of plutonium, was prepared and certified for plutonium and uranium amount content and isotopic abundance. The amount content of the spikes is such that no dilution of a typical sample of dissolved fuel from a reprocessing facility is needed before the measurement by IDMS. The preparation and the certification of the new batch, IRMM-1027n, are discussed in more detail in the certification report [5].

3. Experimental

3.1. Preparation of IRMM-1027n

High purity metals were chosen as starting materials for the IRMM-1027 LSD series. Plutonium MP2 metal (98 % ^{239}Pu) from Cetama, natural uranium (EC NRM 101) and highly enriched ^{235}U metal (NBL CRM-116) were dissolved in concentrated nitric acid in a 3 L long-necked borosilicate flask.

Approximately 1200 units were dispensed into penicillin vials using 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 [5, 6].

3.2. 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·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·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·L^{-1} nitric acid to obtain the final concentration of ^{242}Pu of 0.1 mg Pu per g solution.

For preparation of the mixed plutonium-uranium IRMM-046c spike, ^{233}U metal was dissolved in 8 mol·L^{-1} nitric acid and purified by anion exchange. The uranium fraction was eluted with 8 mol·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·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.

3.3. 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 [4]. The purified fractions of uranium and plutonium were prepared in 1 mol·L^{-1} nitric acid and loaded on a Re filament. The isotopic ratios of uranium and plutonium were measured on a Triton TIMS (Thermo Fischer Scientific) using the total evaporation technique [7, 8]. With 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.

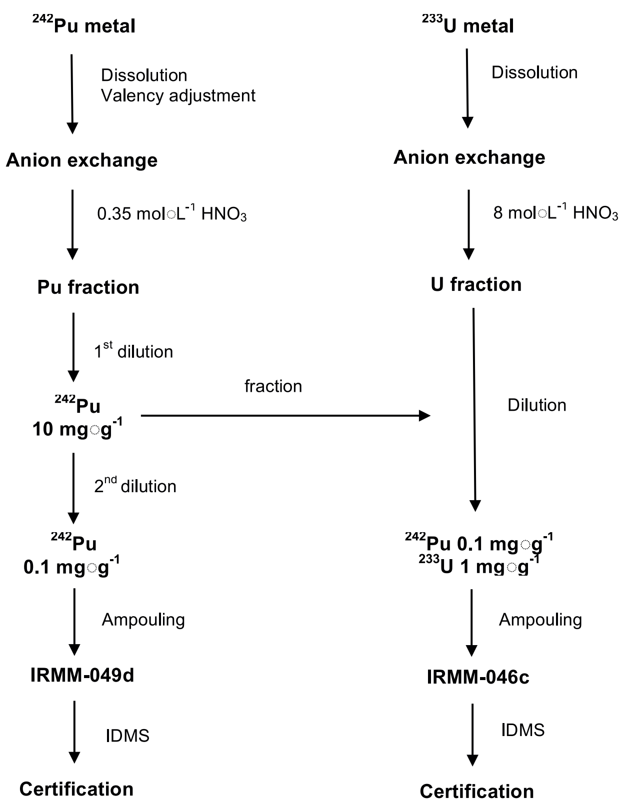

Figure 1: The flowchart for the preparation of IRMM-049d and IRMM-046c.

4. Results and Discussion

4.1. IRMM-1027n

The certification of the 1027 LSD series is done by gravimetry. This allows the isotopic contents of the LSD spike to be certified based on the certificates of the metals (chemical purity and isotopic abundance), the masses of the metals and the solution. As a result the certified values of the uranium and plutonium isotopic contents have small uncertainties. In addition, the certified values of amount contents and isotopic composition 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 assess the homogeneity of the whole series (1200 units), four were selected at random to verify the U amount content by IDMS [5]. 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.

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 IRMM-1027 series 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 [4]. 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 a “primary” measurement method, which has proven to provide accurate results 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 IRMM-1027n to the IRMM compatibility study on selected Pu spikes [4]. 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·g^{-1} [5]. The results for the plutonium amount content are also traceable to the SI via MP2 but with two additional steps in the traceability chain

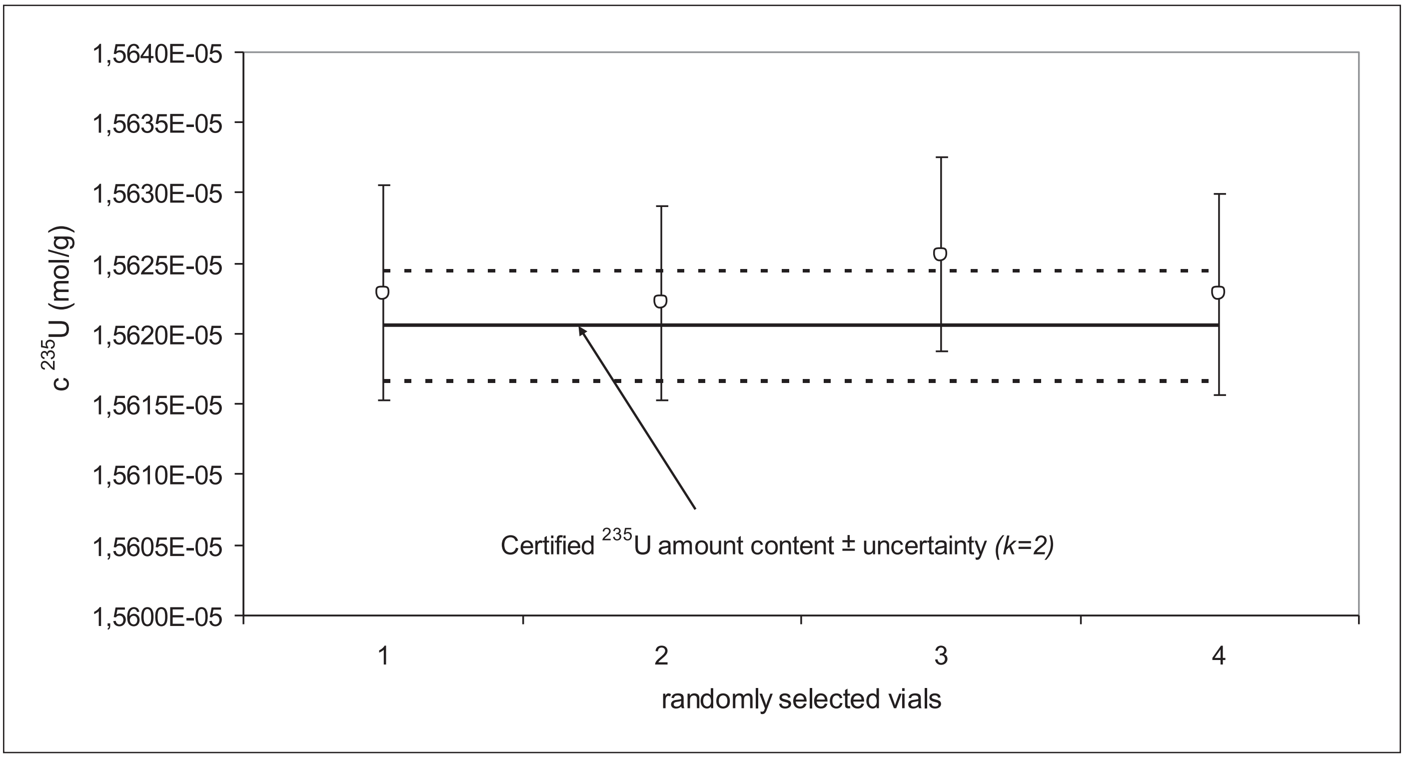

Figure 2: Amount content of ^{235}U in IRMM-1027n (from the masses of metals and solution) compared with the measured values by IDMS (with expanded uncertainties, *k=2*).

IRMM-1027n – via IRMM-046b – via IRMM-1027m (and verified by Eqrain-11) – via MP2 to SI [4]. The Pu amount content in IRMM-1027m was certified by gravimetry via MP2. IRMM-1027m was then used for the re-certification of the IRMM-046b spike reference material by IDMS. Finally, IRMM-1027n was certified by IDMS via the newly certified IRMM-046b. In addition, IRMM-046b was also successfully used as spike reference material for the determination of the plutonium amount content of the external quality control Eqrain-11 certified test sample. In the framework of the ongoing support task EC A 1806, *Verification of mixed U-Pu Spikes,* between IRMM, IAEA and ITU additional verification measurements of IRMM-1027n will be available in the near future.

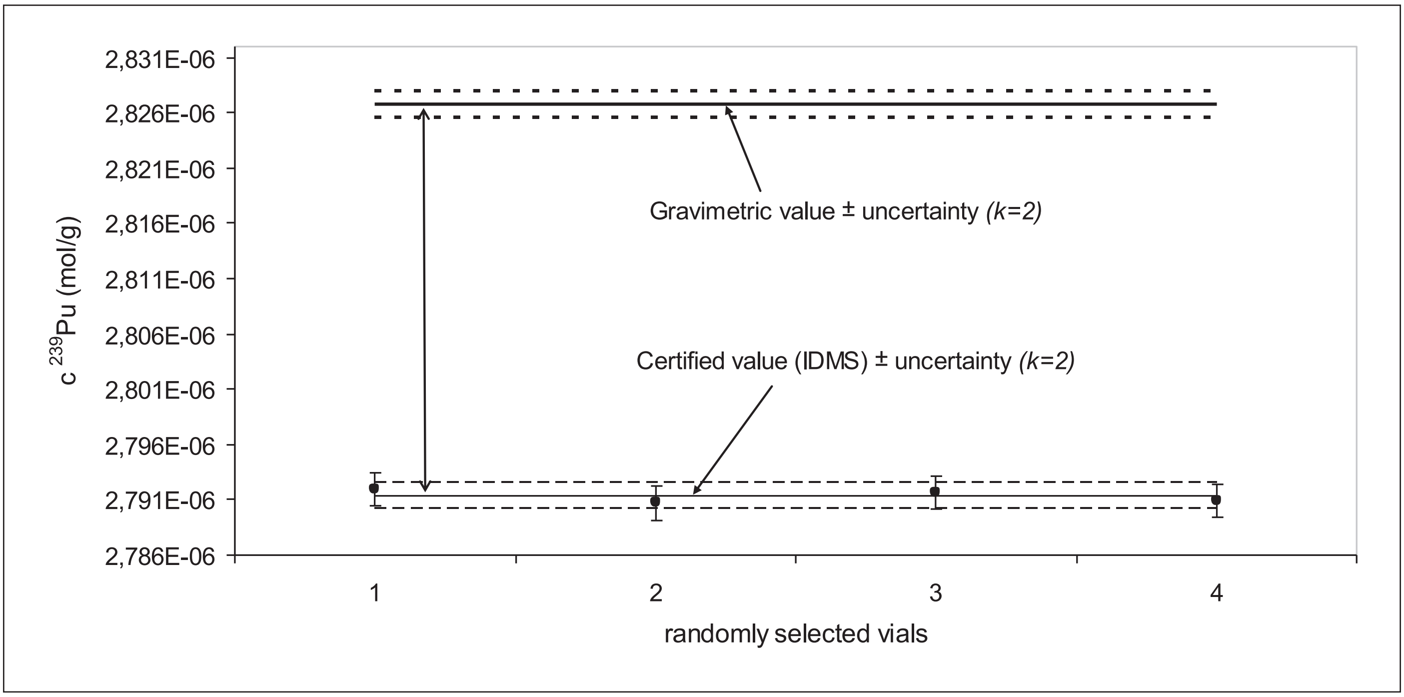

Figure 3: Amount content of ^{239}Pu in IRMM-1027n (from the masses of metals and solution) compared with the measured values by IDMS (with expanded uncertainties, *k=2*).

The individual IDMS results together with the certified value are shown in Figure 3.

4.2. 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.

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. Both of these new Pu CRMs have 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 a 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	0.049	

Table 1: Relative expanded uncertainties of selected spike isotopic reference materials.

4.3 External plutonium inter-laboratory comparison programme Eqrain-11

Eqrain is the inter-laboratory comparison programme organised at regular intervals for the analysis of uranium and plutonium by CETAMA (Commission d'ETAblissement des Méthodes d'Analyse du CEA). 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 [4]. 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 sample M107, normalised to the Eqrain-11 reference value, are shown in Figure 4.

4.4. 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 the 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.

5. 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 and IRMM-046c, mixed uranium-plutonium, and IRMM049d, highly enriched in ^{242}Pu, were prepared and certified for amount content by IDMS. These materials were prepared in the framework 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 par-

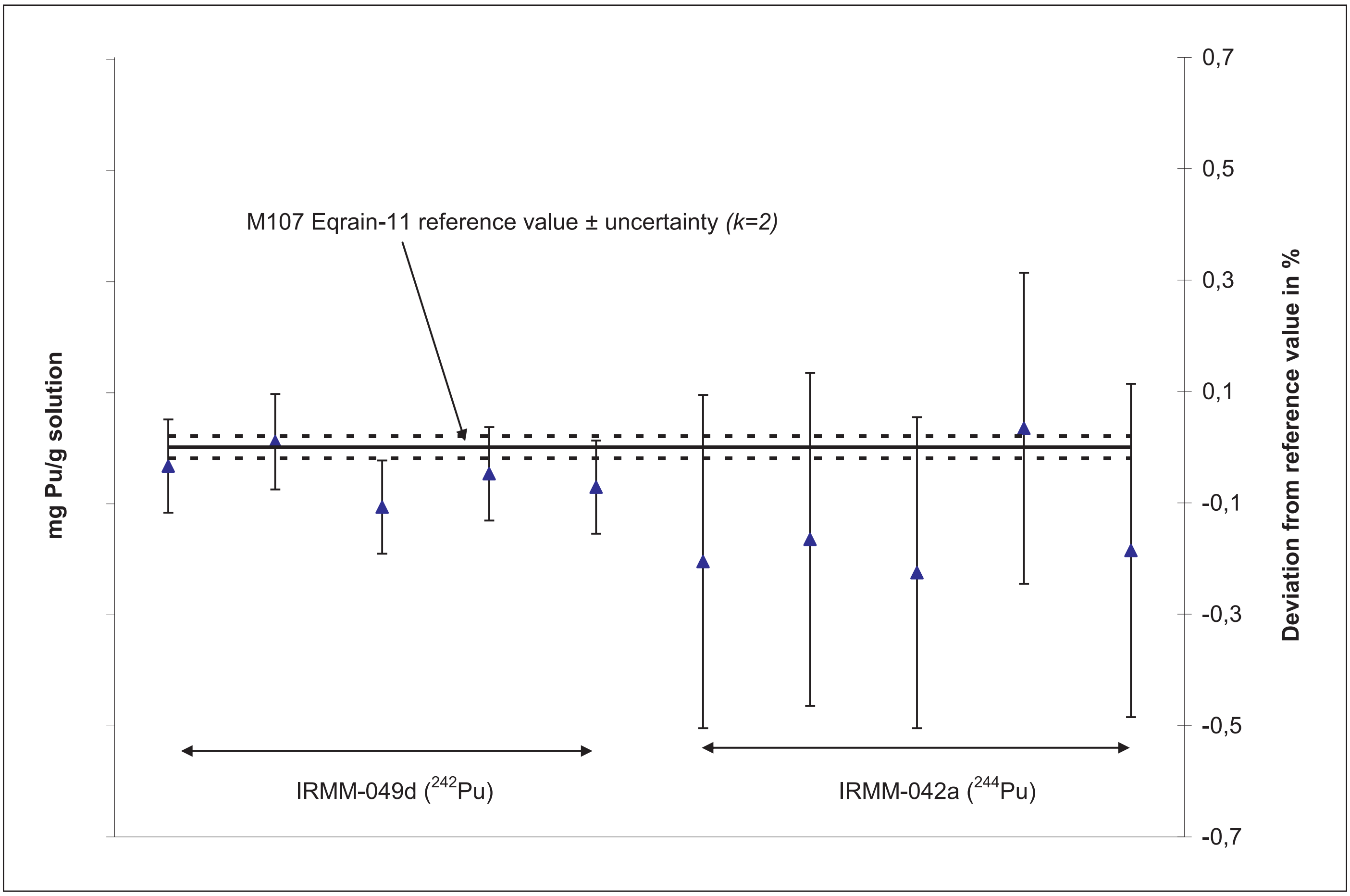

Figure 4: Normalised amount content of ^{239}Pu in Eqrain-11 M107 sample compared with the measured values by IDMS (with expanded uncertainties, *k=2*).

ticipation in Eqrain-11, applying IDMS and using various selected Pu spike 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, using fora such as ESARDA Working Group on Standards and Techniques for Destructive Analysis (WGDA).

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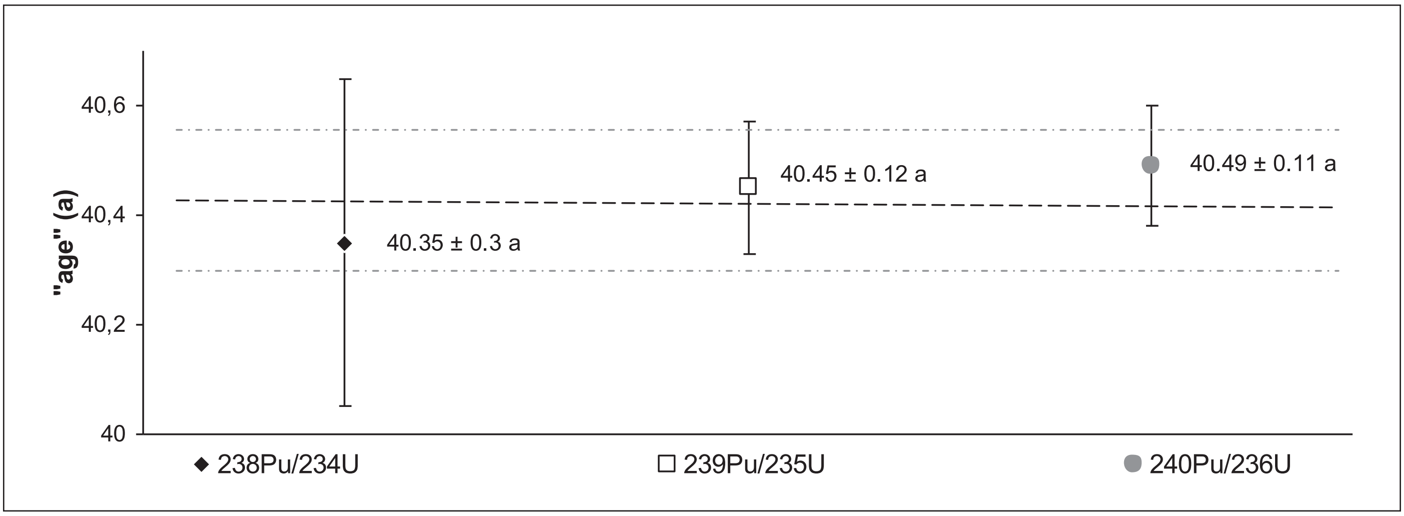

Figure 5: Age in years of NBS SRM 946 (calculated for 17 October 2010) for the “clocks” $^{238}Pu/^{234}U$, $^{239}Pu/^{235}U$ and $^{240}Pu/^{236}U$ with expanded uncertainties *(k=2)*.

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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)

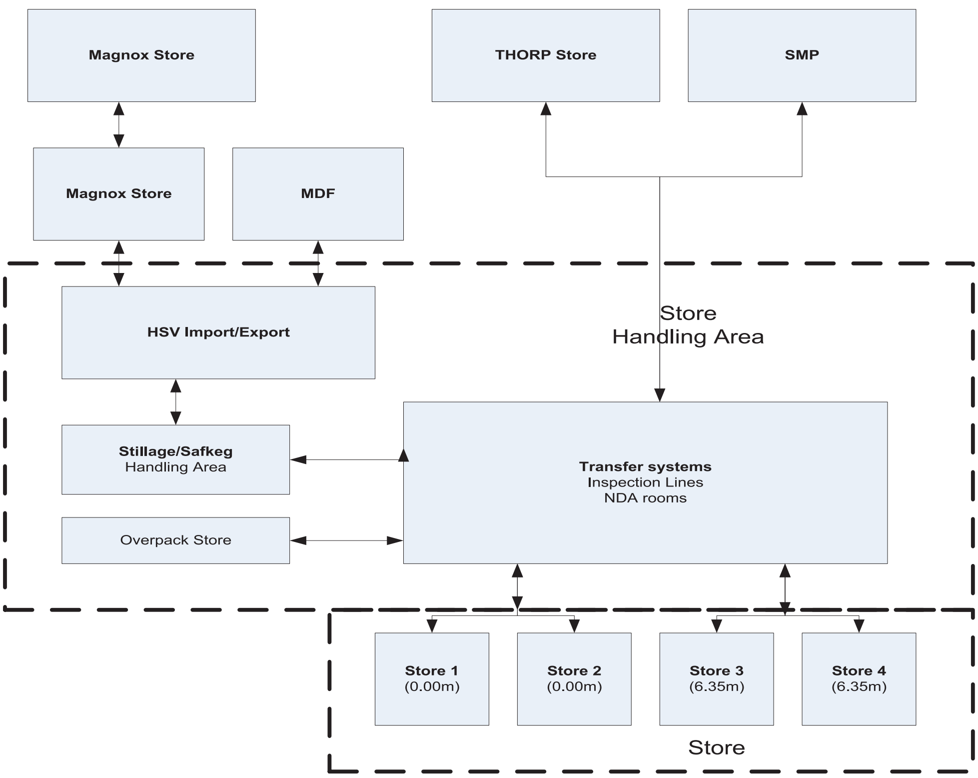

Figure 1: General Scheme for Material Transfers

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 in 2005, 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.

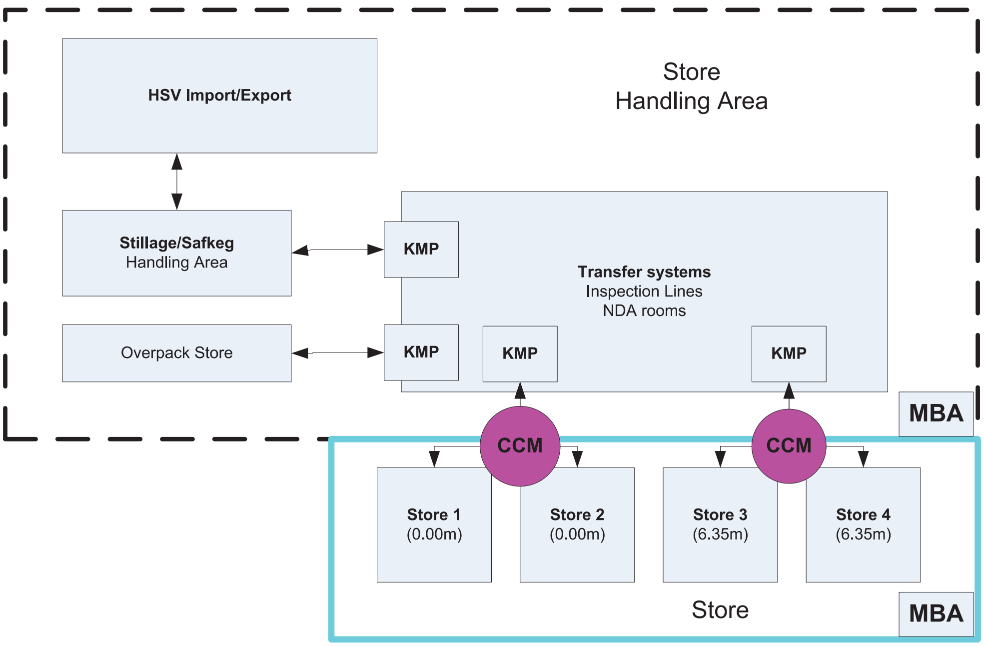

Figure 2: Can flow with NDA KMPs

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.

(Non available due to the sensitive nature of the plant)

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

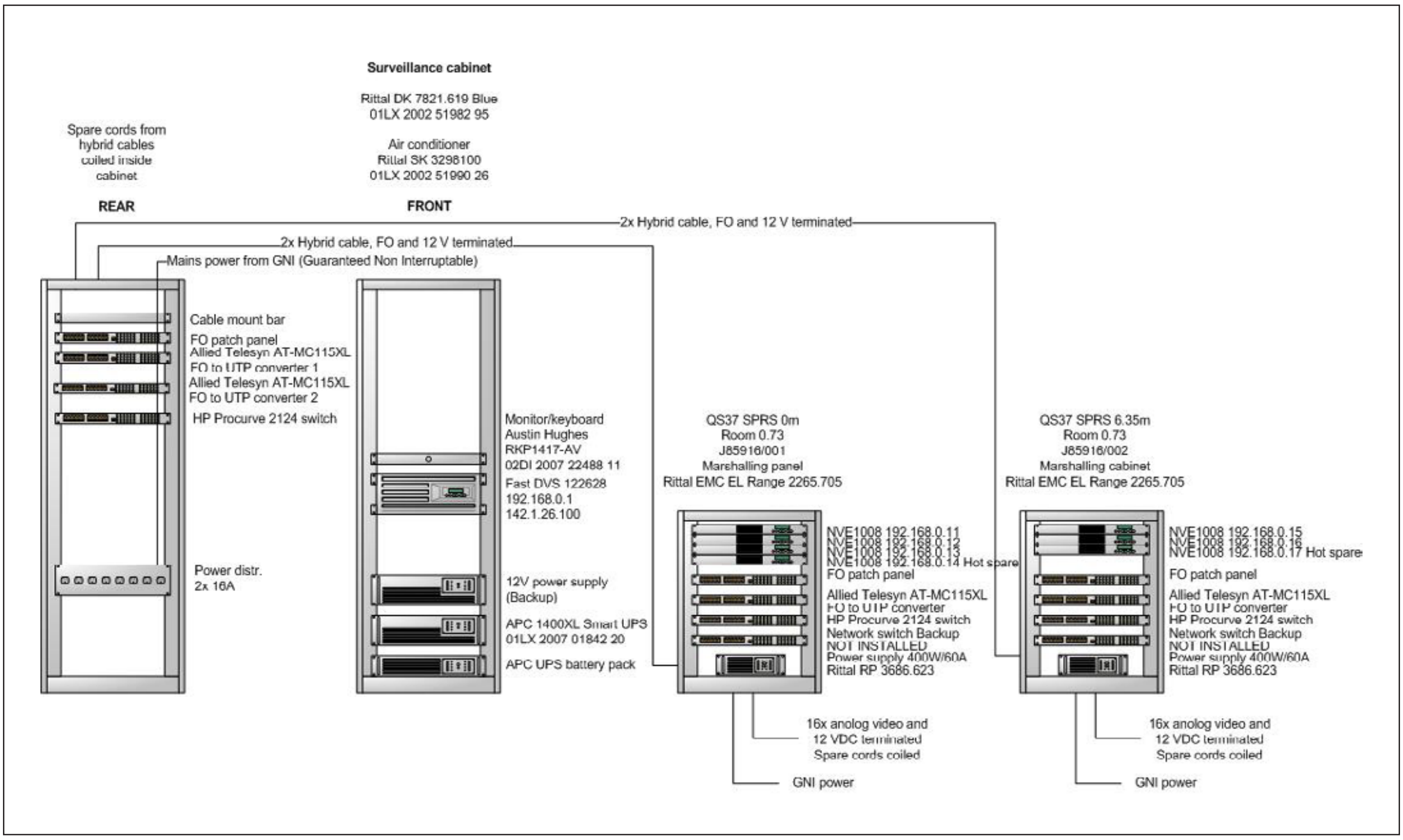

Figure 3: Layout of Surveillance Recording Cabinets within SPRS.

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 imaging 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 demonstrated that there is a willingness to involve the different stakeholders at the initial stages of the project. Although Safeguards By Design is a voluntary collaboration in the future it could well be linked to the requirement to communicate to the Commission, under Article 41 of the Euratom Treaty, any investments in a new project prior to the signature of the first contract.

The outcome of the project has demonstrated that the plant is in an advanced state of design at the 200 days trigger point prior to the start of construction, as detailed in Article 4 of the Commission Regulation 302/2005.The discussions during the design phase emphasised their usefulness to be able to incorporate safeguards measures and instrumentation details at this stage.

The close collaboration of the safeguards inspectors with the designers and plant operators has demonstrated the usefulness of Safeguards By Design 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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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 made over a certain period of time.

The resulting plant design should be a compromise among economical, safety, security and safeguards implementation constraints.

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 . Concerning safeguards, 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 starting from very early design stages would be beneficial for all the involved stake-holders (i.e. the designer , the operator and the national and international regulators).

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 under development facilities with new design features and technologies that require R&D and modifications to the SG approaches, hereinafter recalled.

2. Evolution of Safeguards

All the EU civil nuclear installations are under EURATOM safeguards (Euratom Treaty Article 78). 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 the regulation, which also regulates the information needed to be submitted during the different phases of construction of a new nuclear installation. Although safeguards by design are not legally required to date, they have often been 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 out 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

are aimed at providing assurance that no undeclared nuclear activity is carried inside or outside declared nuclear sites.

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.

Design choices have an impact on safeguards friendliness, but also on non-classical proliferation indicators. In this respect, IAEA's analyses to verify the completeness of declarations take into account also a series of indicators of non-classical indicators like environmental monitoring, satellite imagery, open source information and trade analysis.

The AP requires indeed States to declare their exports of "trigger list" items possibly assisting the establishment of 15 nuclear fuel cycle related activities, as contained in its Annexes.

Figure 1 shows a pictorial view of this approach.

3. The Safeguards by Design process

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 Nuclear Fuel Cycle (NFC), and can serve to replace an old plant, or adding 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 safeguards-by-design process is described e.g. in [3]. The operator launches a call for tender to which suppliers and manufacturers answer with proposed designs and budget. The Regulator will verify the design's compliance with safety standards, as well as security and safeguards requirements [4]. Save the great importance of ensuring the maximum safety and security, regulator and operator must be aware that the less "safeguards-friendly" the design is, the higher is the amount and cost of the later technological adjustments needed to ensure the fulfilment of safeguards objectives.

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 design and tender phase, with an exchange of information between the safeguards inspectorates, the national authorities, the operator and the designer:

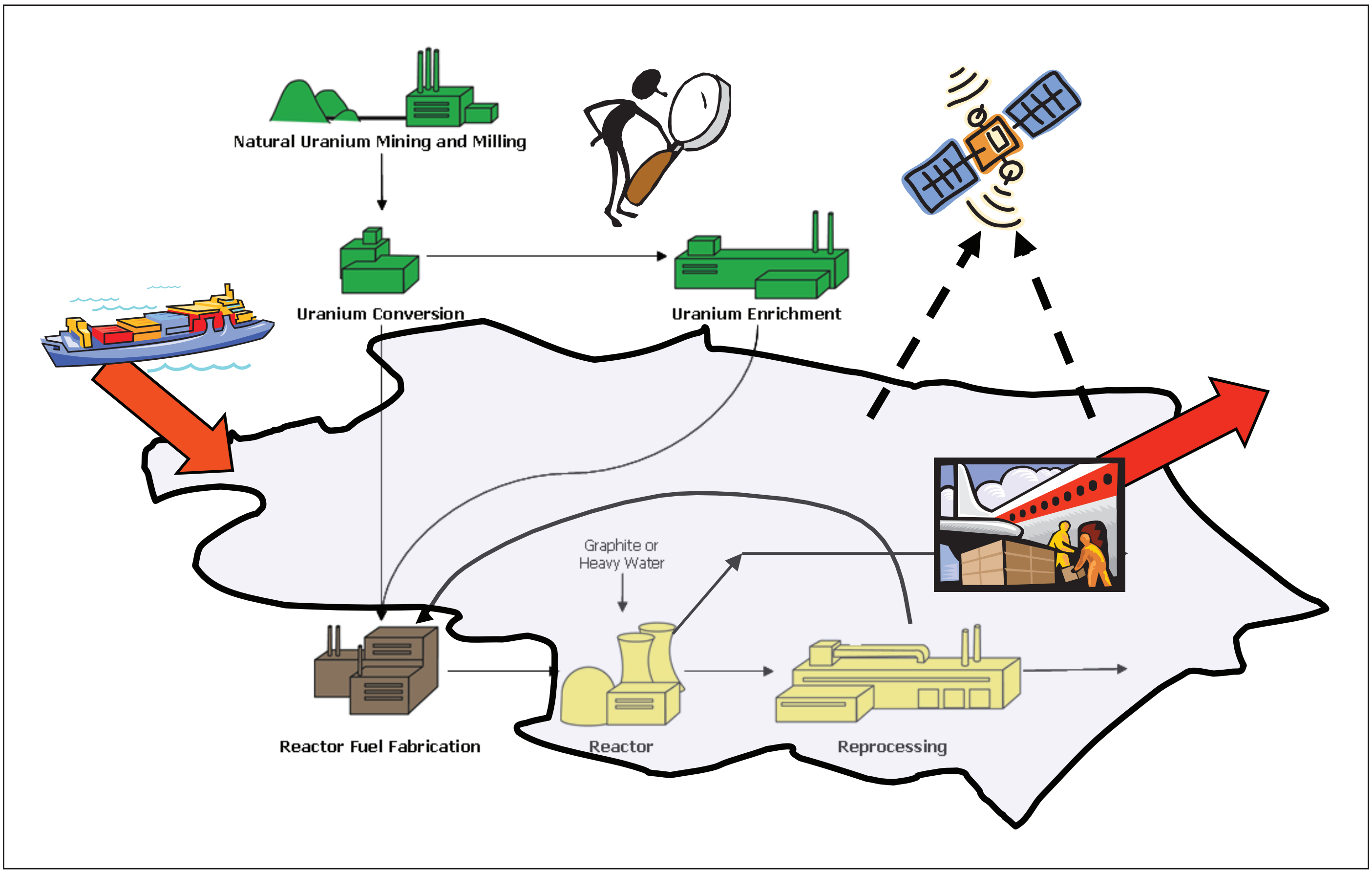

Figure 1: Fully information-driven safeguards

Phases	R/SSAC	IAEA	Operator	Designer
Call for Tender phase	Following decision by Government, provide general information on new facility build to IAEA			
	Identifies operator	Best safeguards practices compilation for the facility type	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 Safeguards guidelines		
Preliminary Design				Preliminary design
			Approval	
	DIQ	Medium level Safeguards guidelines		
	Propose SQ approach	Safeguards approach		
		Detailed SG guidelines		
		Design information evaluation for completeness		
		Feed-back to R/SSAC		
	Feed-back to operator		Feed-back to designer	Feed-back to safeguards equipment supplier
Final design	Draft facility attachment		Approval-submit DI to RSAC/SSAC	Final design
Construction	BTC/DIV verification	DIV Safeguards installation	Safeguards equipment installation	
Commissioning	Final facility attachment	DIV	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	

Table 1: Safeguards by Design process

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.

Tab.1 summarises the milestones and decision steps following the decision to build a facility of certain general characteristics and output. Each step implies safeguardability consequences and R&D needs. 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.

5. A phased approach to SBD: the Safeguardability Check-List

As shown in Fig. 2, the safeguardability attributes can be seen grouped in three categories corresponding to the classical safeguards system components, and also in three phases 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. Each feature in each layer may affect one or more safeguards components.

First phase: Design basic features intrinsic to the process

It includes design features intrinsic to the process with direct proliferation resistance and safeguardability relevance. Their definition relates to the pre-conceptual design phase of Tab. 1.

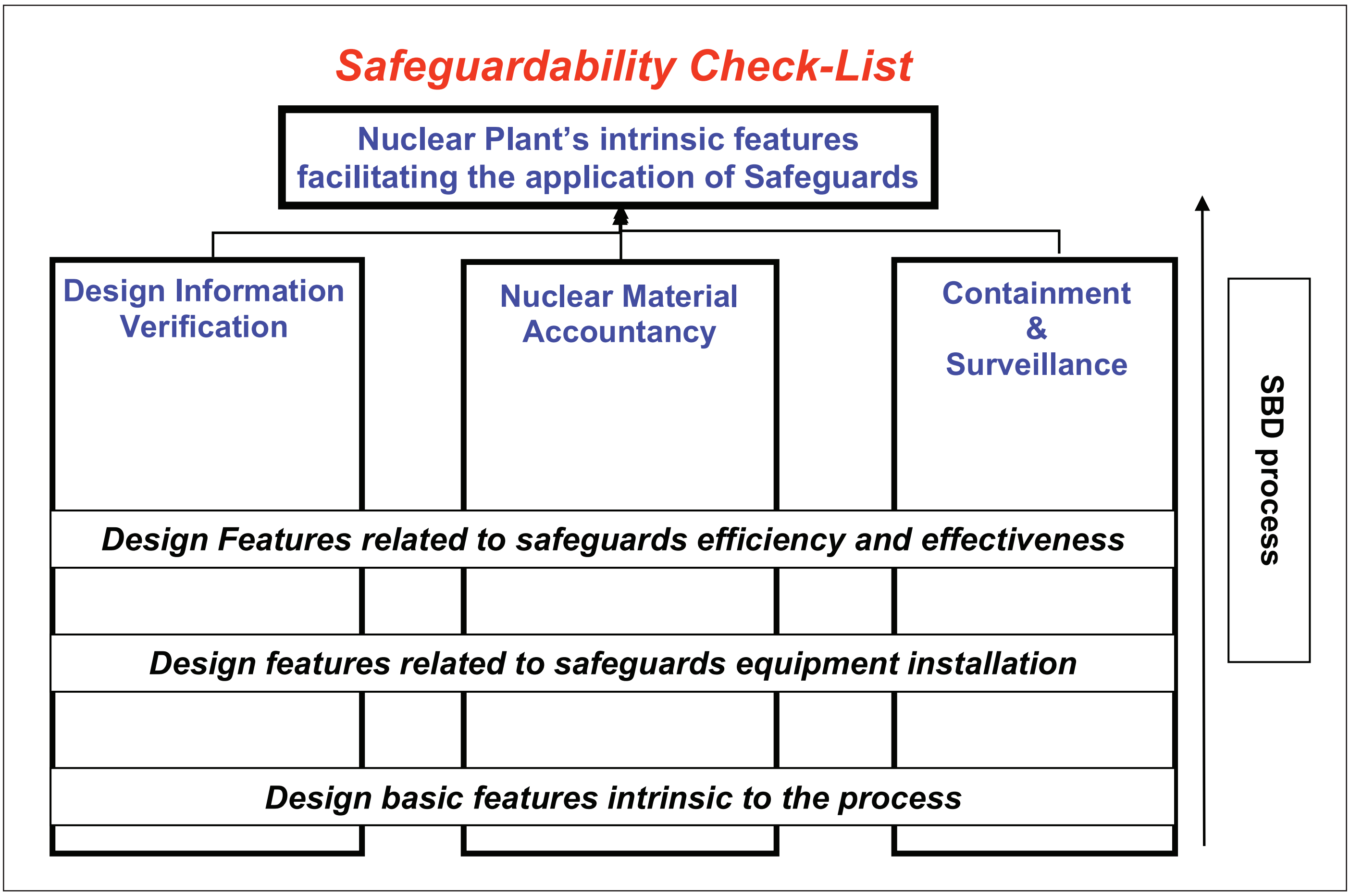

Figure 2: The Safeguardability Check-List model

The basic attributes 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 to a proliferator 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 phase: Design features related to safeguards equipment 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, possibility of visual surveillance and inspection or use of 3D laser reconstruction 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.

Joint-use equipment and remote data transmission should also be taken into account.

6. Discussion

The Appendix contains a non-exhaustive list of SCL attributes building upon existing references and some newly introduced ones.

An example application of SCL to the design of an off-load existing reactor was presented in ESARDA 33rd Symposium, and is thus available in the proceedings.

Each attribute has a suggested metrics in the range between 0 – 1, directly proportional to the estimated safeguardability relevance. The metrics is defined so that at increasing values corresponds a higher associated safeguardability.

> *Example: at increasing level of automation, the possibility of remote monitoring and thus the safeguardability will also increase*

Each attribute can impact more than one safeguards component, and intervene at different phases of the SBD process.

The effect of some attributes (e.g. extent of access to the core, or type and burn-up of the fuel) could also be estimated by means of *utility functions*, correlating their influence on safeguardability. Where available, they are referenced to. Expert elicitation could also be used if no utility function or other estimate is available.

Following the check-list, one can interactively assign safeguardability value to each attribute based on expert judgment or estimates by available utility functions.

After performing the assessment the designer can see where the safeguardability is strong and where it has weaknesses. Some of the attributes are fixed or 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.

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, at the same time maintaining the view of the details. However this would not be a straightforward exercise, requiring the investigation of several parameters.

An overall view of the model, either in tabular form 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 Safeguards-by Design process has been recalled in its ideal development. High level guidelines are being finalised by IAEA based on a contribution by the EC Support Programme.

A set of facility specific Guidelines should then be developed to assist the SBD process.

The main message 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 non-exhaustive list of attributes suggested in the Appendix is open for comments and suggestions. It can be expanded or modified. At the same time, the proposed metrics can be improved, and where applicable more utility functions can be introduced or created.

To facilitate the use of the Safeguardability-Check-List, the attributes relative to the various types of nuclear materials circulating in the facility could be pre-calculated, in order to establish datasets that could be easily used for qualitative parametric studies by Stake-holders.

The next step proposed for the improvement of SCL is to verify the relevance of the proposed attributes and metrics with a detailed SDB demo study, supported by simulation of the safeguards system efficiency and effectiveness.

A further more sophisticated fine tuning of the Safeguardability Check-List model could also try to identify and assess the interactions among attributes.

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APPENDIX: Definition of "Safeguardability Check-List" attributes

1. Nuclear Material Throughput

The nominal NM throughput, is measured in [SQ/year] to put in relation to the safeguards effort. A 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 an inventory of NM. The inventory is measured in significant quantities. In general, 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 typical 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 most "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 $0.2{\cdot}M$ 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.

This parameter is discussed in [9] and [10]

5. Concentration of NM

Higher concentration of materials will be more attractive since a lower mass (or volume) of material would need to be diverted or stolen to acquire a SQ of NM. The metric uses the number of SQ 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 a weighted average should be used for concentration assessment.

As discussed 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.

See also [11] p. 13-14.

7. Frequency of outages (for reactors)

The frequency of outages determines the frequency of the core opening. This requires a bigger safeguards effort by inspectors. Also the reliability of outages according to the scheduled inspection planning should be taken into account.

8. Fraction of processes involving items

In terms of nuclear material accounting, items are easier and normally cheaper to verify than NM 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 a location which for certain reasons cannot be accessed by an inspector. NM Inventory is determined by NM flow from/to this area, hence 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. The integrity and authenticity of data must of course be guaranteed [2].

13. Measurement uncertainty

In bulk handling facilities there may be differences between book inventory and physical inventory due to measurement uncertainty and to 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 in verifying the content of spent fuel pool. The other methods are more time consuming and intrusive. Water quality is hence an 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 labelling in such a way that the ID will be readable [7].

19. NM Holdup

In a nuclear material bulk handling facility there may be locations and areas generating build up of nuclear material arising from the processing operations. These locations need to be clearly identified and the estimated build up quantities calculated. NM's holdup and its fluctuation should be as little as possible [2].

20. Homogeneity

For bulk handling facilities inhomogeneity can significantly contribute to measurement uncertainties because the amount of NM is often determined from small sample. Another example is when 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 the 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 the application of C/S [6].

25. Layout facilitating MBA and KMP definition

The facility should be laid out so that its physical structure and boundaries can be mapped into MBAs and KMPs could be 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. Occasionally sealing of openings could be requested [7].

27. Autonomousillumination? 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 applicability

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 wire 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 RCAS or IAEA's 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, might expand 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, or BTC for EURATOM) 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 detect 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 compared to 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].

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 inspectorates. This paper does not address the participation of the IAEA in the Commission NMAC audit activities. It is not covered the use that the IAEA could do of the audit results. However, the IAEA will benefit of the improvements that the audit activities will bring to the NMAC systems of nuclear operators under common safeguards.

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.

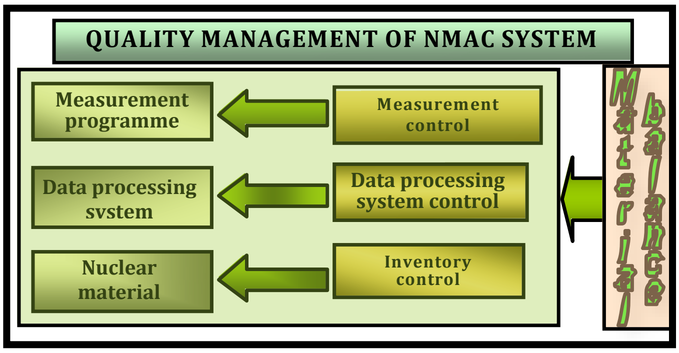

Figure 1: NMAC system model

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.

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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 [1] 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 <u>all</u> information available to the IAEA about the State, both facility-specific factors <u>and</u> 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 [2].

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 [3]. 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 all 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 [4]. 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 <u>differentiation</u> of approaches, not <u>discrimination</u>. 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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[3] 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."

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

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 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,[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 the subject of nuclear export controls is gaining the attention of an increasing number of academics. Notwithstanding the fine quality of these mentioned sources, what is currently lacking 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 first part of the paper presents the reasoning 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 meas-

[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.

ures, 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

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 demands greater legal control.[2] 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.[3]

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.

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."[4] 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.

The Resolution 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.[5] Surely "appropriate and effective" in this case means 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. 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

2 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.

3 Heupel, Monika. "Implementing UN Security Council Resolution 1540: A division of labor strategy." *Carnegie Papers*, No. 87, June 2007.

4 Craft, Cassidy. "*Brief challenges of UNSCR 1540: Questions about international export controls*." Center for International Trade and Security Briefs, March 2009.

5 Bergenas, Johan. "*A piece of the global puzzle*." Stimson Center Report, December 9, 2010.

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 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.

Of course, the reports are not the only tool used in this best practices study. The 1540 Committee 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. In addition, 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. This leaves the 1540 reports and matrices as one of the most valuable sources of information regarding specific national laws and the way that Resolution 1540 is implemented on a national level. It is the matrices in particular that will be the focus of the following sections.

3. Using the 1540 Committee Matrices to Develop a Best Practices Model

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 instrument 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 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."[6]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.

4. Description of the Matrices

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.[7] 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.

The 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 fi-

[6] Crail, Peter. "Implementing UN Security Council Resolution 1540: A risk-based approach." *Non-proliferation Review, Vol. 13, No. 2, July 2006.*

[7] The matrices are contained in worksheets downloadable at http://www.un.org/sc/1540/1540matrix.shtml

nance 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.

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 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.[8] 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.[9] These components of nuclear export controls illustrate the complexity of successful implementation.

5. Border Controls

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.[10] 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.[11] 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.[12] In case radioactive material is found, a system must be in place for radiological emergency response, which involves not just equipment but also the thorough training of personnel.

Border controls also must consider the difficulty in detecting dual use equipment which may contribute to nuclear weapons proliferation. Dual-use goods are difficult to detect because they have civilian applications as well as military ones, making detection of dangerous cases extremely challenging. The detection of dual-use trade should involve more than border equipment, but also the targeting of

8 Stoiber, Carlton; Baer, Alex; Pelzer, Norbert; Tonhauser, Wolfram. IAEA Handbook on Nuclear Law. Vienna: International Atomic Energy Agency, 2003.

9 Number 27 of the matrix is "other"

10 Ibid.

11 This calls into question the role of border controls in both export control and illicit trafficking. Clearly these two activities are linked and strong border controls contribute to curtailing nuclear weapons proliferation through either form. Indeed, licensing officials and customs officers must communicate and cooperate to the furthest extent possible.

12 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).

shipments, Finally, for both military and dual-use goods, 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.[13] 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.

6. Licensing

The second set of measures listed in the 1540 matrices involves licensing procedures. If a domestic export control system is to work effectively, a thorough licensing system should be organized and enforced to control the trade of dual use 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.[14] 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 a 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 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.[15]

A well-working licensing system should account for the difference between individual, general, and global 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."[16] 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.[17] 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, and this is called a global license. Usually global licenses are granted in cases where large amounts of material are being traded between countries working together on defense projects, for example. Exemption from licensing also refers to these situations and by and large covers exports conducted by government bodies and their related contractors and subcontractors.[18] Export control provisions should account for

13 Gabulov, I.A. "Emerging nuclear security issues for transit countries," in *Radiation Safety Problems in the Caspian Region*. Amsterdam: Kluwer Academic Publishers, 2004.

14 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!

15 Harding, Margaret. "Spaghetti with meatballs: Nuclear export control reform." *The Energy Collective*. October 14, 2010.

16 Licensing Unit, UK Export Control Organization, August 3, 2009.

17 Export regulations, customs benefits and tax incentives." United States Commerce Department, Chapter 11.

18 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]

such exemptions by unequivocally stating in which cases exports do not need a license.

Two additional 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.[19] 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.

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."[20] 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.[21] 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.[22]

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 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.[23] These controls are especially difficult to implement in a globalized world where information exchange takes place very often among specialists from dif-

19 "Assistance for foreign atomic energy activities." United States Atomic Energy Act Section ,57b.

20 Gates, Robert. "Export control reform." (Presented at the Business Executives for National Security, April 20, 2010, Washington DC).

21 Harding, Margaret. "Spaghetti with meatballs: Nuclear export control reform." *The Energy Collective*. October 14, 2010.

22 Beck, Michael. "Reforming the multilateral export control regimes." *The Nonproliferation Review*, Vol. 7, No. 2, Summer 2000.

23 "The deemed export rule in an era of globalization." United States Department of Commerce, December 20, 2007.

ferent 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.

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 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.

7. 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[24] 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. The list produced by the Nuclear Suppliers Group and published as INFCIRC/254, is divided in two parts: Part 1, or "Trigger List"[25], and Part 2 related to dual use equipment. The Trigger 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 so-called "EU dual use control list" contained in Annex I to EC Regulation 428/2009 assigns a category to materials and equipment

[24] 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.

[25] i.e. equipment "triggering" the need for international safeguards

depending on what they can be used for in a non-nuclear context, except for Category 0, which lists nuclear material, facilities and equipment all taken from the above mentioned Trigger List (INFCIRC 254/Part 1). All the nuclear dual use items taken from INFCIRC 254/Part 2 are instead distributed in Categories 1-9 dedicated to special materials, material processing, electronics, computers, telecommunications and "information security", sensors and lasers, navigation and avionics, marine items, and aerospace and propulsion equipment.[26]. The EU list was developed in the 90's as a first attempt to integrate into one document the controls from the four international regimes plus the Chemical Weapons Convention. It was later adopted by various non-EU countries as well as by the US, which included about 25% more controls, maintaining the same structure and coding.

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."[27]

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 was well-informed of the items that should be controlled, perhaps there would be no need for specific lists at all. Notwithstanding this point, control lists are extremely important 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 not just based on 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 information necessary for 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

[26] 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.

[27] 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.

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.[28] 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.[29] 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 further element of a comprehensive control list, as specified likewise by the 1540 Committee matrix, is the inclusion of end-user controls. It should be reiterated that these are one measure not explicitly specified in the 1540 Resolution, but rather considered a part of effective export control implementation by the 1540 Committee. 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.[30] 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 it 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.[31]

Several models for catch-all clauses exist in various nuclear export control laws. INFCIRC/254/Rev.8/Part 2 of the NSG guidelines is in this case one of the clearest models for such a clause. The guideline states that “Suppliers should ensure that their national legislation requires an authorization for the transfer of items not listed in the Annex if the items in question are or may be intended, in their entirety or in part, for use in connection with a “nuclear explosive activity.” Article 4 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 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.”[32] 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

[28] 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.

[29] 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.

[30] Pietsch, Georg. “End-user and end-use controls.” (Presented at the 9th International Export Control Conference, Dubrovnik, Croatia, 20-22 October 2008).

[31] Robdrup, Dothe. “*Catch-all.*” Danish Enterprise and Construction Authority, 2010.

[32] Ibid.

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.[33] 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.[34]

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 applicable to open source information.[35] 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.[36] 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."[37] 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. For non-members of Regimes, 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

[33] "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).

[34] "Strategic Commodities Control System Trade and Industry Department Frequently Asked Questions." Strategic trade control Hong Kong website, http://www.stc.tid.gov.hk/.

[35] "Best practices for implementing intangible transfer of technology controls." Drafted at the Wassenaar Arrangement Plenary Meeting, Stokes Australia, December 2006.

[36] "Controls on tangible and intangible transfers of technology." United States Office of Export Control Cooperation, Website: www.exportcontrol.org

[37] Strategic Goods Control Act, Singapore Customs Authority, January 1, 2003.

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.

8. 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 provide several 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 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."[38] Trans-shipment occurs when such goods **do not** leave the stated environments, as it typically happens in Free Trade Zones. 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.[39]

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. 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.[40] 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

[38] UK Export Control Act, 2002.

[39] "*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.

[40] The Financial Times Lexicon, http://lexicon.ft.com/

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."[41] 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 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.

9. 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.[42]

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.

10. Information

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.

[41] 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).

[42] 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.

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, to raise awareness. 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.[43] 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 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.[44]

11. 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.[45] 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 legally-binding 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 succeed. 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 a rubric from which to measure

[43] Hund, G and Seward, A. "*Broadening industry governance to include non-proliferation.*" Pacific Northwest Center for Global Security Report." November 11, 2008.

[44] Beck, Michael. "Reforming the multilateral export control regimes." *The Nonproliferation Review*, Vol. 7, No. 2, Summer 2000.

[45] (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)

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.

Group Representation of the Prompt Fission Neutron Spectrum of ^{252}Cf

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Abstract

We review the spectral representation used for the prompt fission neutron spectrum of ^{252}Cf in the International Organization for Standardization document ISO 8529-1. We find corrections to Table A.2, the discrete group structure form, of this report are needed. We describe the approach to generating replacement values and provide a new tabulation.

Keywords: NDA; Monte Carlo; Prompt Fission Neutron Spectrum, ISO 8529

1. Introduction

Increasing use is being made of sophisticated Monte Carlo neutron modeling to predict the performance and extend the calibration of neutron assay systems for safeguards [1]. It is necessary to evaluate and report defensible uncertainties associated with such computations that are fit for purpose. The field of data evaluation tells us that workers often underestimate uncertainty [2]. In the evaluation of potential sources of bias associated with Monte Carlo neutron modeling, sensitivity analysis is an important assessment tool. In the case of calculating the detection efficiency to ^{252}Cf fission neutrons, as an example, one might undertake calculations using various representations of the spectrum. The true spectrum is not known of course since it is subject to experimental and data evaluation uncertainty. Because of this it is important to calculate the impact on the desired results that variations to the input spectrum cause. The variations should represent the uncertainty in the state of the knowledge of the true spectrum or reflect the consequences of picking one procedure over another. Standardization is beneficial in that allows different workers to use common data sets of fundamental physical properties without having to create their own reference values. In performing a sensitivity study one can therefore simply select from different reputable evaluations or recommended consensus standards in order to get a reasonable estimate on how strongly data uncertainties affect the end results.

In our work we turned to ISO 8529 [3] as a readily assessable and widely used reference to the representation of the ^{252}Cf Prompt Fission Neutron Spectrum (PFNS). A functional form and also a histogram form are presented in [3]. We found the table to be inconsistent with the algebraic form as we shall discuss.

2. Review of the ISO 8529 Representation

The energy distribution of the neutron source strength, $B_E(E)$, is approximated by the following formula:

$$B_E(E).dE = B.\frac{2}{\sqrt{\pi}}.\sqrt{\frac{E}{T}}.e^{-E/T}.\frac{dE}{T}$$

where, $B_E(E).dE$ is the number of neutrons per second emerging with energies in the incremental interval dE about energy E. Integrating over all neutron energies, $E = 0\ to\ \infty$, shows $B_E(E)$ is normalized to B, the neutron source strength in neutrons per sec. When the neutron energy E is measured in units of MeV the spectrum parameter T takes the value of 1.42 MeV [3].

For convenience we recast the spectral distribution in natural energy units and renormalize to unity:

$$\chi(x).dx = \frac{2}{\sqrt{\pi}}.\sqrt{x}.e^{-x}.dx$$

where the dimensionless energy parameter $x = E/T$ and runs from $0\ to\ \infty$.

The function $\chi(x)$ is normalized to unity and $\chi(x).dx$ is the probability that a neutron will emerge within the differential interval dx about the value x. $\chi(E)$ is zero at both $x = 0$ and $x = \infty$. When $x \ll 1$ $\chi(E)$ varies as $\frac{2}{\sqrt{\pi}}.\sqrt{x}$ while, at the other extreme of the range, when $x \gg 1$, $\chi(E)$ rapidly approaches zero from above in a way that the gradient vanishes. In between the function peaks at $x = \frac{1}{2}$. The mean value of $x = \frac{3}{2}$. For ^{252}Cf with $T = 1.42$ MeV this corresponds to a mode energy of 0.71 MeV and a mean energy of 2.13 MeV.

To create a histogram, or group structure, from the algebraic spectral distribution we must define the group boundaries, x_i, and have a means of integrating between then. Thus, we arrive at the following expression for the group contents – that is the probability that a neutron will emerge having energies in multiples of T between x_i and x_{i+1}:

$$\chi_i = \int_{x_i}^{x_{i+1}} \chi(x).dx = \int_0^{x_{i+1}} \chi(x).dx - \int_0^{x_i} \chi(x).dx = [Y(x_{i+1}) - Y(x_i)]$$

where we have made explicit use of the cumulative probability function given by the following:

$$Y(\theta) = \int_0^{\theta} \chi(x).dx = \mathrm{erf}(\sqrt{\theta}) - \frac{2}{\sqrt{\pi}}.\sqrt{\theta}.e^{-\theta}$$

In this expression $\mathrm{erf}(z)$ is the error function defined by the integral:

$$\mathrm{erf}(z) = \frac{2}{\sqrt{\pi}}.\int_0^{z} e^{-t^2}.dt$$

Evaluation of the χ_i for a given set of x_i is now straightforward since all the terms involve functions that are readily

Index *i*	E_i MeV	*x* E_i/T	Cumulative Integral, Y_i	Contents χ_i This Work	Contents χ_i ISO Table A.2	Difference %
0	0	0.00E+00	0.0000E+00	1.18E-10	*Not Listed*	-
1	4.14E-07	2.92E-07	1.1838E-10	3.26E-10	3.10E-10	-5.20
2	1.00E-06	7.04E-07	4.4450E-10	1.36E-08	1.11E-08	-22.64
3	1.00E-05	7.04E-06	1.4058E-08	1.43E-07	1.27E-07	-12.69
4	5.00E-05	3.52E-05	1.5717E-07	2.87E-07	2.76E-07	-4.12
5	1.00E-04	7.04E-05	4.4454E-07	8.13E-07	7.82E-07	-3.93
6	2.00E-04	1.41E-04	1.2573E-06	2.30E-06	2.21E-06	-4.01
7	4.00E-04	2.82E-04	3.5559E-06	4.68E-06	4.53E-06	-3.20
8	7.00E-04	4.93E-04	8.2309E-06	5.82E-06	5.68E-06	-2.49
9	1.00E-03	7.04E-04	1.4052E-05	5.89E-05	5.51E-05	-6.90
10	3.00E-03	2.11E-03	7.2956E-05	1.33E-04	1.28E-04	-4.01
11	6.00E-03	4.23E-03	2.0609E-04	2.37E-04	2.30E-04	-2.87
12	1.00E-02	7.04E-03	4.4269E-04	8.04E-04	7.74E-04	-3.90
13	2.00E-02	1.41E-02	1.2468E-03	2.25E-03	2.17E-03	-3.69
14	4.00E-02	2.82E-02	3.4970E-03	2.87E-03	2.80E-03	-2.63
15	6.00E-02	4.23E-02	6.3705E-03	3.36E-03	3.29E-03	-1.99
16	8.00E-02	5.63E-02	9.7260E-03	3.75E-03	3.68E-03	-1.98
17	1.00E-01	7.04E-02	1.3479E-02	1.08E-02	1.05E-02	-2.58
18	1.50E-01	1.06E-01	2.4250E-02	1.23E-02	1.21E-02	-1.78
19	2.00E-01	1.41E-01	3.6566E-02	1.35E-02	1.33E-02	-1.41
20	2.50E-01	1.76E-01	5.0053E-02	1.44E-02	1.42E-02	-1.40
21	3.00E-01	2.11E-01	6.4451E-02	1.51E-02	1.49E-02	-1.43
22	3.50E-01	2.46E-01	7.9564E-02	1.57E-02	1.55E-02	-1.12
23	4.00E-01	2.82E-01	9.5238E-02	1.61E-02	1.60E-02	-0.69
24	4.50E-01	3.17E-01	1.1135E-01	1.64E-02	1.63E-02	-0.87
25	5.00E-01	3.52E-01	1.2779E-01	1.67E-02	1.66E-02	-0.54
26	5.50E-01	3.87E-01	1.4448E-01	1.69E-02	1.68E-02	-0.37
27	6.00E-01	4.23E-01	1.6134E-01	3.40E-02	3.38E-02	-0.59
28	7.00E-01	4.93E-01	1.9534E-01	3.40E-02	3.39E-02	-0.42
29	8.00E-01	5.63E-01	2.2938E-01	3.38E-02	3.37E-02	-0.23
30	9.00E-01	6.34E-01	2.6316E-01	3.33E-02	3.33E-02	0.05
31	1.00E+00	7.04E-01	2.9644E-01	6.44E-02	6.46E-02	0.27
32	1.20E+00	8.45E-01	3.6087E-01	6.09E-02	6.12E-02	0.57
33	1.40E+00	9.86E-01	4.2173E-01	5.68E-02	5.73E-02	0.89
34	1.60E+00	1.13E+00	4.7852E-01	5.25E-02	5.31E-02	1.09
35	1.80E+00	1.27E+00	5.3104E-01	4.82E-02	4.88E-02	1.16
36	2.00E+00	1.41E+00	5.7927E-01	6.46E-02	6.55E-02	1.43
37	2.30E+00	1.62E+00	6.4384E-01	5.58E-02	5.67E-02	1.58
38	2.60E+00	1.83E+00	6.9965E-01	6.22E-02	6.33E-02	1.71
39	3.00E+00	2.11E+00	7.6186E-01	6.11E-02	6.21E-02	1.60
40	3.50E+00	2.46E+00	8.2297E-01	4.62E-02	4.68E-02	1.33
41	4.00E+00	2.82E+00	8.6915E-01	3.46E-02	3.49E-02	0.92
42	4.50E+00	3.17E+00	9.0373E-01	2.57E-02	2.58E-02	0.35
43	5.00E+00	3.52E+00	9.2944E-01	3.30E-02	3.30E-02	-0.01
44	6.00E+00	4.23E+00	9.6244E-01	1.78E-02	1.74E-02	-2.06
45	7.00E+00	4.93E+00	9.8020E-01	9.44E-03	9.01E-03	-4.76
46	8.00E+00	5.63E+00	9.8964E-01	4.97E-03	4.61E-03	-7.84
47	9.00E+00	6.34E+00	9.9461E-01	2.60E-03	2.33E-03	-11.58
48	1.00E+01	7.04E+00	9.9721E-01	1.35E-03	1.17E-03	-15.56
49	1.10E+01	7.75E+00	9.9856E-01	7.00E-04	5.83E-04	-20.04
50	1.20E+01	8.45E+00	9.9926E-01	3.61E-04	2.88E-04	-25.30
51	1.30E+01	9.15E+00	9.9962E-01	1.85E-04	1.42E-04	-30.62
52	1.40E+01	9.86E+00	9.9981E-01	9.51E-05	6.94E-05	-36.99
53	1.50E+01	1.06E+01	9.9990E-01	9.91E-05	*Not Listed*	
54	∞	∞	1.0000E+00			
Sums				1.0000	1.0027	

Table 1: Group representation of the ^{252}Cf fission spectrum using the bin structure of ISO 8529 and the integration scheme described in the main text.

computed. In our case we simply used the standard functions available in the popular spreadsheet software MS EXCEL®.

3. Results

In Table 1 we recreate Table A.2 of [3] adopting the same group structure and algebraic spectral representation for the ^{252}Cf PFNS but using the integration scheme just described in place of the unspecified numerical integration referred to in [3]. In addition we list the cumulative probability distribution and the difference between the ISO group contents and the current results. We find significant and variable differences between the two histograms suggesting a problem with the numerical approach used in [3].

4. Conclusions

In using the ISO 8529 recommended neutron spectrum for ^{252}Cf we found the histogram representation to be in error. Using analytical integration rather than the original unspecified numerical integration we have generated replacement values for Table A.2 of reference [3]. The significance of this work is due to the fact that [3] is a widely used source of reference spectra for instrument modeling and blind adoption of the histogram could inadvertently mislead users.

Acknowledgements

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[3] International Organization for Standardization (2001), Reference neutron radiations – Part 1: Characteristics and Methods of Production, ISO 8529-1:2001-02-01(E), see also ISO 8529-1:2001/Cor 1:2008(F) which provide a typographical correction to Table A.4 the ^{241}Am-Be(α,n) spectrum.

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 UF6 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 UF6 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. Miller et al. previously showed modelling results for the expected signatures from product, feed, and tails cylinders. [6] Experience with the UCAS in Japan has shown that the Singles increase linearly with uranium mass. For product cylinders, the concept is to use the Doubles-to-Singles or cadmium ratio to give a measure of the ^{235}U enrichment. Simulations have shown that both the Doubles-to-Singles and cadmium ratio increase linearly with ^{235}U enrichment. Field trials of the system will help determine if one of the signatures is more suitable for cylinder assay than the other. For example, we expect that the cadmium ratio may be more useful in measurement scenarios where the background is significant because it exploits a more localized effect (i.e., the thermal neutron albedo) than the Doubles-to-Singles ratio. For the case of feed and tails cylinders, simulations have also shown that the cadmium ratio is a more useful signature than the Doubles-to-Singles ratio for determining enrichment. Again, simulations have shown that the cadmium ratio increases linearly with enrichment in 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 3He tubes. There are three output sig-

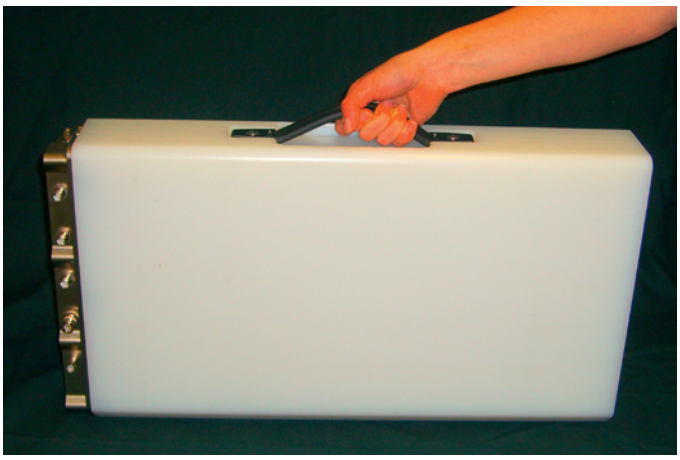

Figure 1: PNEM detector pod.

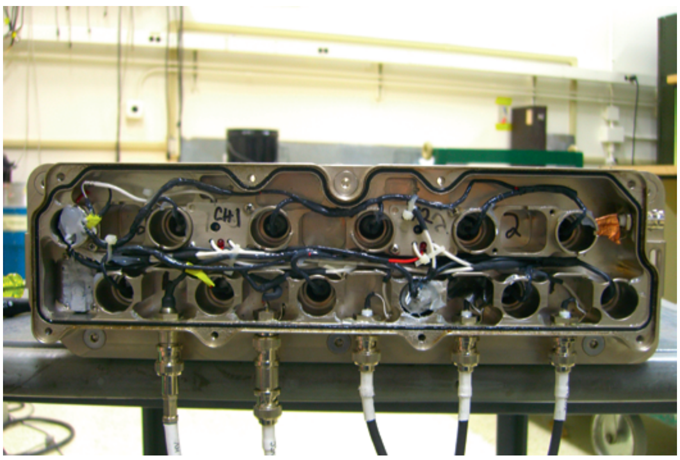

Figure 2: PNEM electronics package.

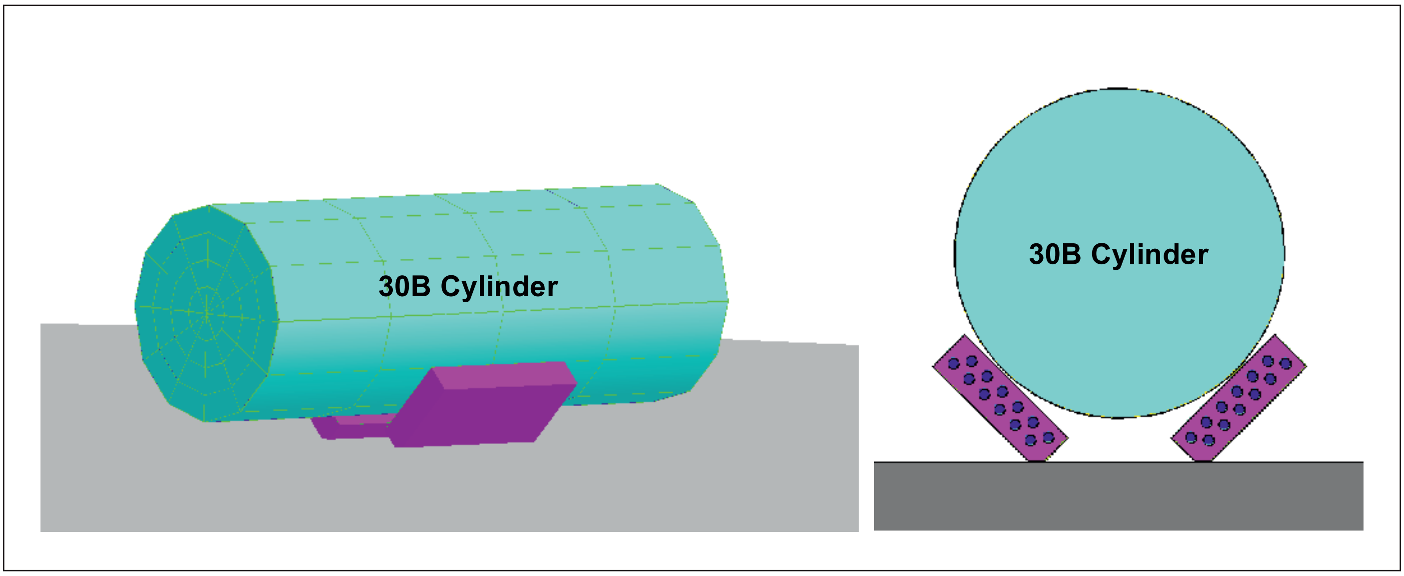

Figure 3: PNEM measurement position with respect to a 30B cylinder.

nals 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.

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]

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. We expect a full 30B cylinder containing low-enriched UF_6 to have a Singles count rate (S) of about 10,000 cps and a Doubles count rate (D) of about 200 cps. The following equation is used to determine the statistical uncertainty on the Doubles:

$$\frac{\sigma_D}{D} = \frac{\sqrt{D + 2GS^2}}{D\sqrt{t}}$$

where G is the gate width (64 μs) and t is the count time in seconds. Using this equation, a typical 30B cylinder will achieve 1-2% statistics in about 20 minutes with the 4 atm prototype system. If the ^{3}He 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 UF6 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.

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 UF6 inside 30B and 48Y cylinders.

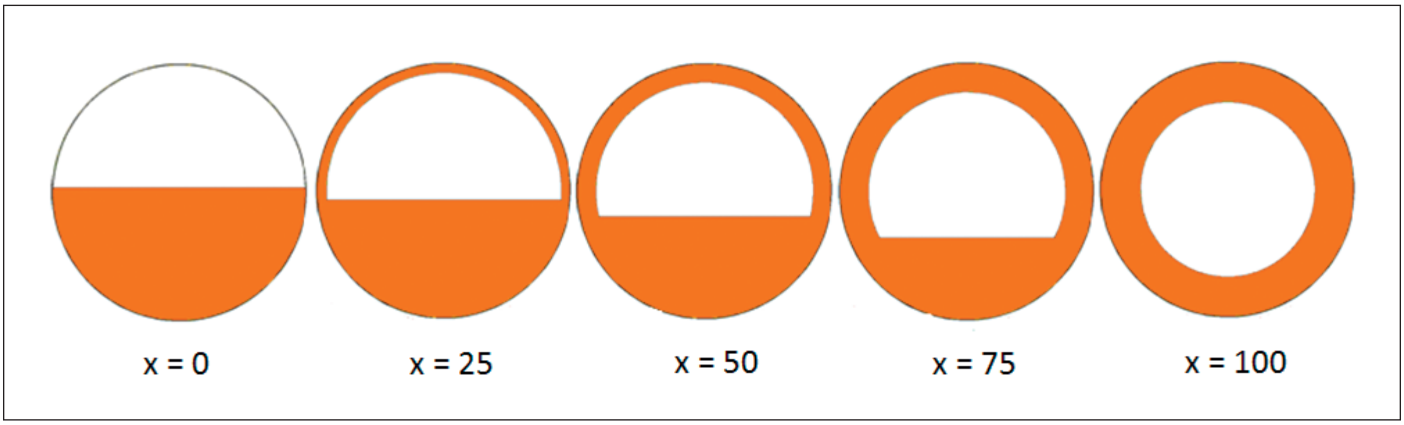

Figure 4: UF_6 filling profiles.

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.

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 3He 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.

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 D1 and D2 and gate widths G1 and G2, where G2 is twice G1, using the following equation:

$$\tau = -\frac{G_1}{ln(D_2/D_1 - 1)}$$

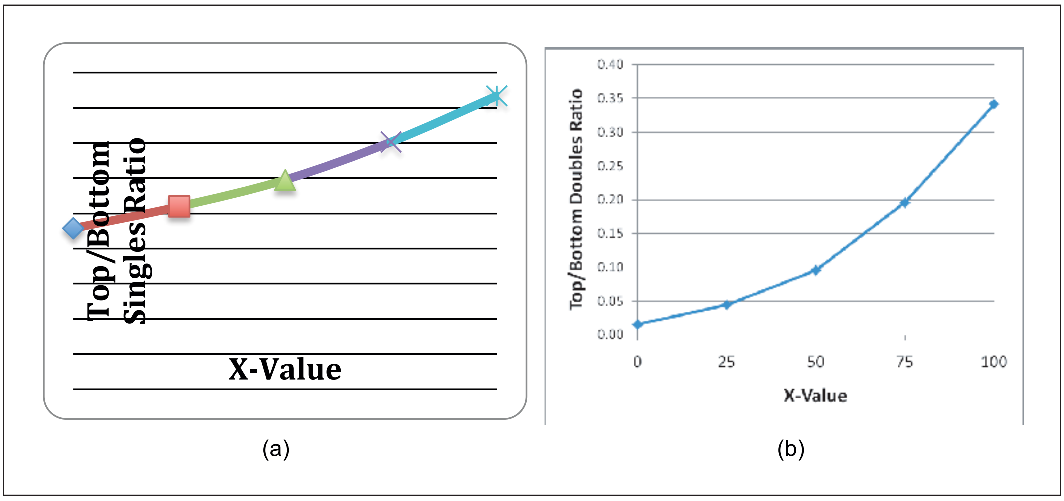

Figure 5: Ratio of top-to-bottom pods as a function of the x-factor for (a) singles and (b) doubles count rates.

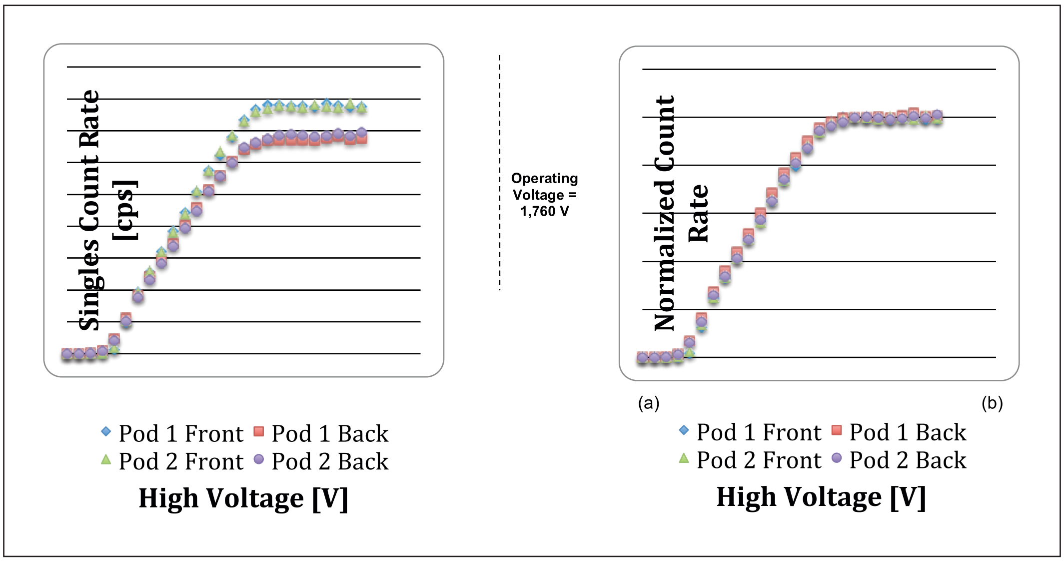

Figure 6: PNEM voltage plateaus showing (a) absolute measurements and (b) normalized count rates.

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 UF6 cylinder measurements, we also calculated the deadtime parameters using the twin source method.

The last series of measurements was performed to characterize the detector response profiles and benchmark the MCNPX calculations. Again, we used a 252Cf 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.

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.

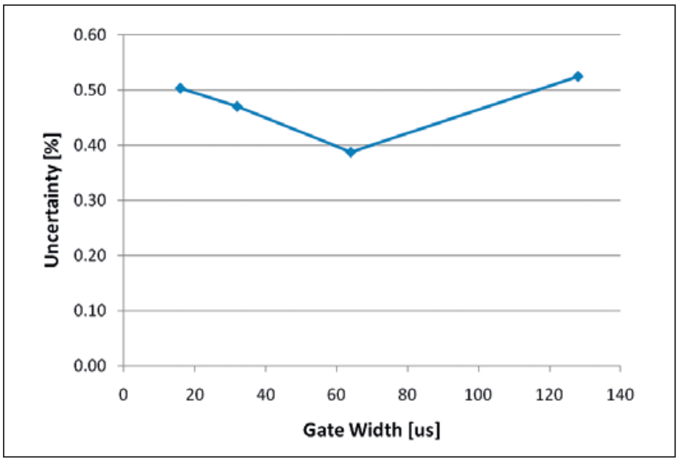

Figure 7: Percent uncertainty in the doubles count rate as a function of gate width.

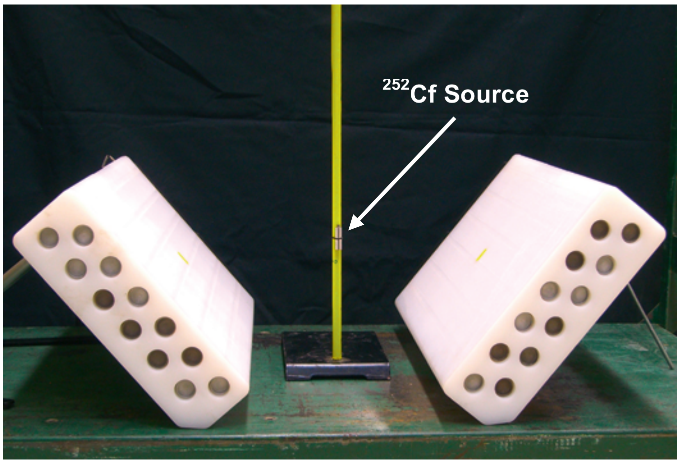

Figure 8: Photograph of the PNEM detector pods in the proposed 30B measurement position.

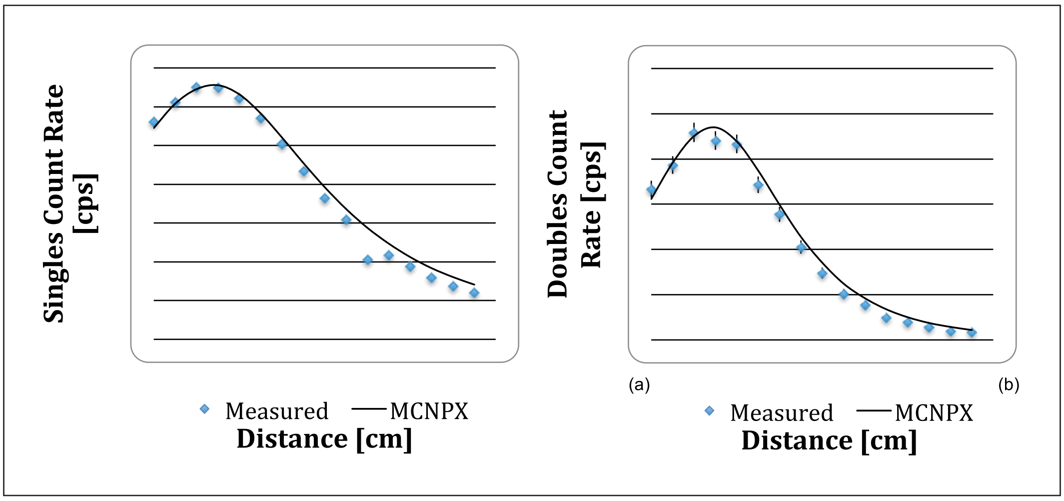

Figure 9: Measured and calculated response profiles for the (a) singles and (b) doubles count rates.

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 UF6 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 UF6 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

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Working Groups activities

Verification Technologies & Methodologies WG

Verification Technologies & Methodologies WG

Michel Richard, VTM WG Chairman

Along 2010 and 2011, the activities of the VTM working are strongly marked by two activities. First, the work of the 2010 Reflexion Group, the outcomes of which are essential for ESARDA to adapt its activities and operation in a changing international and European context to support its members, the EU institutions, the IAEA and other organisations for the implement nuclear safeguards and combat of proliferation. Second, the preparation of the triennial 2011 INMM ESARDA joint workshop on forward-looking topics in the area of international safeguards and non-proliferation which will address *"Future Directions for Nuclear Safeguards and Verification"* to be held in Aix en Provence in October.

Management Team/Organisation: Rudolf Avenhaus (Scient. Secret.), Louis Victor Bril (Secretariat), Michel Richard (Chair), Gotthard Stein (Vice-chair). To better address the wide scope of issues dealt with in the VTM three subgroups has been created:

- On Environmental Monitoring (Chair: Dr. Martin Kalinowski).
- On Satellite Imagery (Chair: Dr. Bhupendra Jasani)
- On export control (Chair: Dr. Filippo Sevini)

Members: Participants to the VTM meetings come from many institutions and bodies , on ad hoc basis: they are (non exhaustive list). Members of the Other ESARDA WG (C/S, NDA, DA,..), EC DG (as appropriate), EUSC, European Laboratories, IAEA: Safeguards Departement, Office of Nuclear Security, Export control authorities, Satellite imagery laboratories

Mission Statement and Terms of Reference: The mission of the *ESARDA Working Group on Verification Technologies and Methodologies* is to provide the safeguards 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. To this end the VTM Working Group has been charged with the following tasks:

- 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.

In the future, the mission Statement and the terms of Reference of the VTM will have to be slightly changed to adapt with those of the new group NA/NT which has been created in 2010. Some overlaps of activities between different ESARDA Working Groups does not raise any diffi-

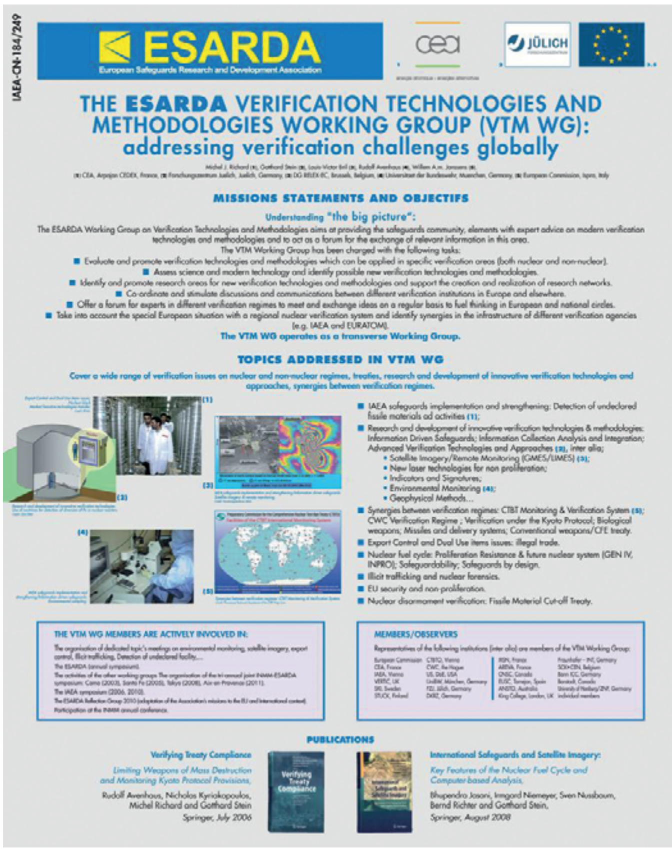

Figure 1: Poster on the VTM objectives and activities.

culties as the scope and the approaches of the issues are different, they complement each other.

Scope of VTM activities: Since its inception in 2002 VTM activities addressed a wide scope of topics related to: Safeguards implementation, Non proliferation, Nuclear security, Disarmament verification, Treaty verification, Technologies/detection equipment R &D: how to use it?, Method of implementation/inspection: verification & monitoring, Legal and institutional aspect,

Topics already addressed by the VTM (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

Activities 2008 (for memories): Remote environmental sampling for nuclear safeguards meeting, Luxembourg (May 2008), Convenors Gotthard Stein and Martin Kalinowski;

Participation to INMM-ESARDA joint meeting, Tokyo, October 2008, along with C/S WG and DA WG, TKM

Products

- 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.
- INMM and ESARDA Symposium Presentations & articles, ESARDA Bulletin article

Activities 2009: ESARDA Seminar, Vilnius (May 2009) : VTM internal meeting and contribution to the ESARDA seminar, INMM annual Conference , Tucson (July 2009), ESARDA outreach (with other WG ðW. Janssens), LIMES Workshop, EUSC/Torrejon (October 2009: Validation of the integrated framework and platform supporting the Non-Proliferation image analyst by integrating data and documents from multiple sources). VTM Fall meeting, JRC/Ispra (November 2009)

Activities 2010: ESARDA internal seminar, Luxembourg (May 2010), Joint session with other WG as C/S DA & NDA and deal with specific topics;; INMM annual conference, Baltimore, (July 2010): Presentation ESARDA/VTM objective and activities; IAEA safeguards symposium, Vienna (1-5 November 2010);

- **2010 Reflexion Group,** 7 meeting in 2010: Objective: provide report and recommendation which allows ESARDA to adapt to the new context & challenges
- **Organisation of the joint 2011 INMM ESARDA meeting, Aix en Provence October 2011**

VTM objectives for 2011 and beyond

- Support IAEA Safeguards objectives; Early detection of non compliance; Information driven safeguards
- Support EU security objectives: EU strategy against WMD proliferation; Support EU external security policy; Instrument for Stability; GMES; CBRN task force; Definition & Implementation of non-proliferation, disarmament, environment Treaties; NPT; CTBT ⇨ EIF? ⇨ OSI specific verification protocol) & IMS; support Cut-off : negotiation inception?; Disarmament verification: decommissioning of facilities/disposition of nuclear material; Environment ⇨ Post Kyoto?
- Improve coordination with other ESARDA WG, and EU Think tanks (Vertic, IPFM, Insap, ...) and INMM.
- Update the ESARDA web site VTM WG page ⇨ Tools of communication between members / Archives of the past

R&D Activities of the ESARDA Novel Approaches / Novel Technologies Working Group (NA/NT WG)

H. Taivonen NA/NT WG Chairman

In recognition of the continuing evolution of the nuclear industry and the need for evermore efficient and effective safeguards tools in support of emerging and future nuclear treaty verification activities, ESARDA established the Novel Approaches / Novel Technologies Working Group (NA/NT WG) under the Chairmanship of Mr. Harri Toivonen in January 2010. An informal NA/NT WG meeting was organized during the 32nd ESARDA Annual Meeting in Luxembourg (May 2010) to introduce ESARDA members and participants to the new WG and to draw the attention of the scientific community into the discussion of novel technologies that may have potential benefits in support of the implementation of safeguards, nuclear security and the verification of other nuclear disarmament, arms control and non-proliferation international treaties.

Objectives of NA/NT WG

The NA/NT WG provides expert advice and assistance to international nuclear inspectorates on novel approaches and technologies having the potential to improve early detection, efficiencies and effectiveness of inspection, monitoring and verification methods for safeguards, nuclear security and verification of international treaties involving nuclear disarmament, arms control and non-proliferation.

A full copy of the WG's Terms of Reference (ToR) is attached.

Tasks

To achieve the above objectives the NA/NT Working Group will, *inter alia*:

- Advise and assist the European Commission (EC), the International Atomic Energy Agency (IAEA), other international safeguards inspectorates and the safeguards community on novel approaches and technologies that could be developed towards operational applications;
- Assist the development and implementation of novel technologies that meet safeguards implementation needs, particularly those that deter the proliferation of nuclear weapons, by detecting early undeclared activities and the misuse of nuclear material or facilities.

Achievements in 2010

A scientific workshop was convened as a part of the NA/NT WG inaugural meeting in Vienna (October 2010). Ten presentations were delivered, covering various novel topics. In general discussion, the WG foresees the following:

- Growing importance of novel methods and instruments based on optical techniques, like laser-based spectroscopy, optical detection systems and radiation imaging;
- Identification of non-proliferation and verification organization needs;
- Establishment of contacts and collaboration with scientific community on a range of topics;
- Establishment of a mechanism for initiating and evaluating novel technologies by the WG, as well as lines of communication to States' experts on specific WG topics;
- Adaption of arms control and nuclear security to the work agenda.

Achievements in 2011

The following major activities took place in 2011:

- Items of interest to the NA/NT WG were identified;
- Second NA/NT workshop was held in Budapest (20 May 2011);
- Third NA/NT workshop was held in Helsinki together with the NDA WG on stand-off detection technologies (28-29 Sep 2011). Three subgroups were established to work on optical stand-off detection methods; stand-off detection of antineutrinos; and novel methods for the verification of future arms control and disarmament treaties.

Participants

Currently, the NA/NT WG has more than 50 members, associate members, observers or individuals from EURATOM, JRC, IAEA, CTBTO, AWE, NNL, CEA, CNRS, SCK.CEN, STUK, Int. Isotopes, CNSC, US National Laboratories, universities and industry. Experts in specific scientific fields will be invited to the meetings for the assessment and development of novel technical proposals.

Joint activities with other Working Groups

NA/NT WG seeks close cooperation with all ESARDA working groups. Particular collaboration is foreseen with the NDA, DA, C/S and VTM which have R&D items where NA/NT could provide fruitful input and further resources.

Terms of Reference of Novel Approaches and Novel Technologies (NA/NT) Working Group of ESARDA

Objectives

To provide expert advice and assistance to international nuclear inspectorates on novel approaches and technologies having the potential to improve early detection, efficiencies and effectiveness of inspection, monitoring and verification methods for safeguards, nuclear security and verification of international treaties involving nuclear disarmament, arms control and non-proliferation.

Definitions

Novel	Not applied previously to safeguards applications or a new, striking approach to improve existing detection, measurement or analysis methods
Novel Approach (NA)	Solutions not applied previously to safeguard applications
Novel Technology (NT)	Usage and knowledge of tools, techniques, crafts, systems or methods not applied previously to safeguards applications

Besides nuclear sciences, NA/NT refers often to other disciplines. Typical examples are optical measurements, simultaneous utilization of different techniques from different disciplines and related algorithms for data analysis. NA/NT WG seeks close cooperation with other ESARDA working groups. “Novel” is by no means the privilege of NA/NT; the aim is to promote scientific research and development of methods and techniques for safeguards and nuclear security.

Tasks

To achieve the above objectives the NA/NT Working Group will:

1. Advise and assist the European Commission (EC), the International Atomic Energy Agency (IAEA), other international safeguards inspectorates and the safeguards community on novel approaches and technologies that could be developed towards operational applications
2. Develop and maintain a comprehensive list of needs and technical aspirations that can be used to support the implementation of emerging and future non-proliferation verification regimes
3. Assist non-proliferation organizations with the identification and prioritization of appropriate methods and instruments in support of their respective R&D efforts
4. Assist the development and implementation of novel technologies that meet safeguards implementation needs, particularly those that deter the proliferation of nuclear weapons, by detecting early the misuse of nuclear material or facilities
5. Establish and maintain a review of novel approaches and technologies
6. Investigate possible transfer of technology from non-nuclear domains to the safeguards area and promote the exchange of information and experience among inspectorates, safeguards authorities and technology developers
7. Promote synergies between safeguards and nuclear security and foster collaboration with other organizations with similar interests and requirements
8. Collaborate with other ESARDA working groups to share expertise in the development of novel tools for safeguards

The ESARDA Working Group on Containment and Surveillance Activities in 2010

Chairman's Report

João G.M. Gonçalves

1. General Information

The ESARDA Working Group on Containment and Surveillance has 18 members and observers from R&D establishments, safeguards equipment manufacturers, safeguards inspectorates, plant operators, regulatory agencies, and ministries. The following ESARDA organisations are represented:

ESARDA Organisations	
European Commission (DG ENERGY, DG JRC)	German nuclear operators (GNS, VGB)
STUK – Finnish nuclear regulatory authority	German Jülich Research Centre
SSM – Swedish nuclear regulatory authority	NNL – British nuclear laboratory
IRSN – French Institute for Radiation Protection, Safety and Security	British Sellafield Safeguards Department
AREVA – French nuclear industry	CNCAN – Romenia National Commission for Nuclear Activities Control (*)
ENEA – Italy's Agenzia nazionale per le nuove tecnologie, l'energia e lo sviluppo economico sostenibile (*)	

Observers	
IAEA – International Atomic Energy Agency	CNSC – Canadian Nuclear Safety Commission
ABACC – Argentine-Brazilian Safeguards Authority	US DoE - Sandia National Laboratories
ASNO – Australian Safeguards and Non-Proliferation Office	

(*) Joined the C/S Working Group in 2010

In 2010, the working group met twice: (a) a meeting in May at the European Commission premises in Luxembourg, and a meeting in October at JRC, Ispra site, Italy. The following topics were addressed:

- Data Security: impact in C/S instruments, methods and approaches
- Interface between Safeguards and Security
- Containment and Surveillance for Final Disposal Facilities
- EURATOM Requirements for C/S systems under Integrated Safeguards
- IAEA's Next Generation Surveillance System – an update on the development work
- Enhanced Data Authentication System (EDAS) – report from Ispra demonstration and workshop
- Caladiom: Intelligent Camera Technology
- Remotely Monitored Seal Array
- Reflections on the results of the INMM – ORNL C/S Workshop (Oak Ridge, June 2010)

As part of the objective in disseminating the best practices in Safeguards Containment and Surveillance, the working group addressed and prepared the following documents:

- Technical Sheet on Laser Based Design Information Verification (available at the ESARDA website)
- Guidelines for sealing, identification, and containment verification systems (soon to be published)

Recurrent activities include: general information exchange, discussions on current R & D projects, maintaining a web based compendium on C/S instrumentation, support of ESARDA Editorial Committee, Training and Knowledge Management (TKM) Working Group and, more recently, the ESARDA Reflection Group 2010.

2. Progress and Highlights of the C/S Woking Group in 2010:

2.1. Data Security: impact in C/S instruments, methods and approaches

A technical discussion of data security for containment and surveillance (and nuclear material safeguards generally) was moderated by Robert Hutchinson, a data security expert from Sandia National Laboratories, USA, who was invited to the ESARDA May Workshop in Luxembourg. Discussion topics focused on important aspects determining the overall security concept, security policy of the organisation, key management, and public key infrastructure. The importance of having a threat model was stressed. There is a need both for research and for training of personnel. Mr. Hutchinson made a clear distinction between technical approaches to security (e.g., the selection of a cryptographic algorithm) and procedural and policy oriented approaches (e.g., management of cryptographic keys). The role of human factors in data security was highly emphasised. Another discussion focused on the security evaluation of digital systems in safety systems. Declaring a software free of compromising items is impossible. The impact of Commercial Off-The-Shelf products in security was also discussed.

In the case of an Unattended Remote Monitoring System (URMS), data security involves the implementation and management of multiple concepts, such as, data integrity, confidentiality and aunthenticity. Mr. Hutchinson explained how these multiple concepts are implemented in practice. In order to have the potential for new concepts one should not limit the requirements to private key cryptography.

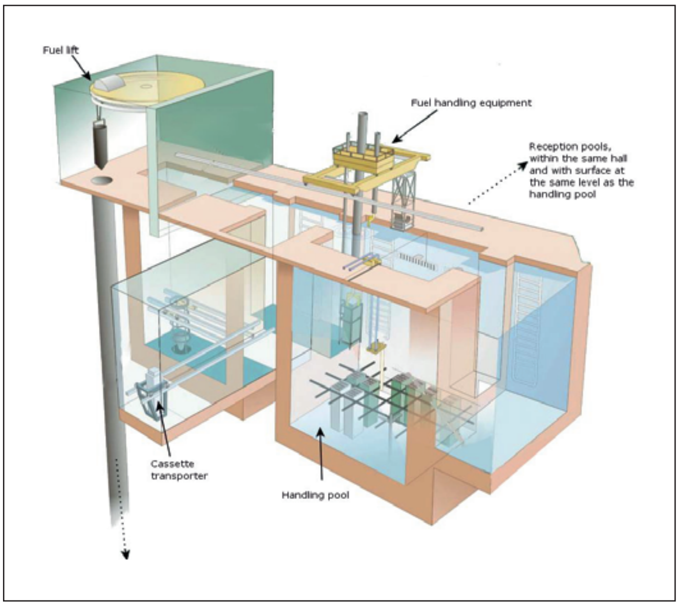

Figure: Sketch of the future Encapsulation Plant for the preparation of spent fuel prior to final disposal (Courtesy of SKB, Sweden)

2.2. Containment and Surveillance for Final Disposal Facilities

The Working Group addressed this topic in both 2010 meetings. There were presentations and updates on the construction of final disposal facilities at Finland, Germany and Sweden. Discussions focused on the prospective Containment and Surveillance methods and devices to be used during the different phases of the preparation of the spent fuel prior to storage as well as any potential mechanisms for later reverification (if required).

Of particular interest are the future containment measures, including identification, authentication and verification of integrity) applied to the canisters with the spent fuel. Different methods and devices were suggested, the vailidity of which depending on extensive practical experimentation and testing. It may happen there is the need to develop new C/S techniques should the existing ones prove inadequate.

A timely definition of the safeguards approach for final disposal facilities by EC's DG-Energy and by the IAEA will contribute to the selection of the corresponding implementation measures.

2.3. Enhanced Data Authentication System (EDAS)

Enhanced Data Authentication (EDAS) is a concept for sharing data from sensors (or instruments) owned by a plant operator with a Safeguards inspectorate, as additional information with potential safeguards relevance. EDAS collects the data as close as possible to the sensor and provides data authentication and encryption such that Safeguards authorities have full confidence in the origin and integrity of the data received.

Practically, EDAS is a box to be inserted between the operator's instrument and the operator's control system, so as to intercept communications in both directions (i.e., data and commands), and to register the copy passed to the inspector branch with additional authentication and encryption. By design, the EDAS box does not influence or interfere with the operator process.

The EDAS team (SNL, JRC and DG-Energy) reported the results of a workshop held at JRC-Ispra where the first EDAS prototype was practically demonstrated to representatives of both IAEA and DG-Energy. During the workshop, the EDAS concept was demonstrated with two industrial sensors:

a) Pressure sensor (Mensor 6180) used in process monitoring applications, and

b) 3D laser based distance measurement system (SICK LMS 200-30106) used in surveillance, safety and other security applications

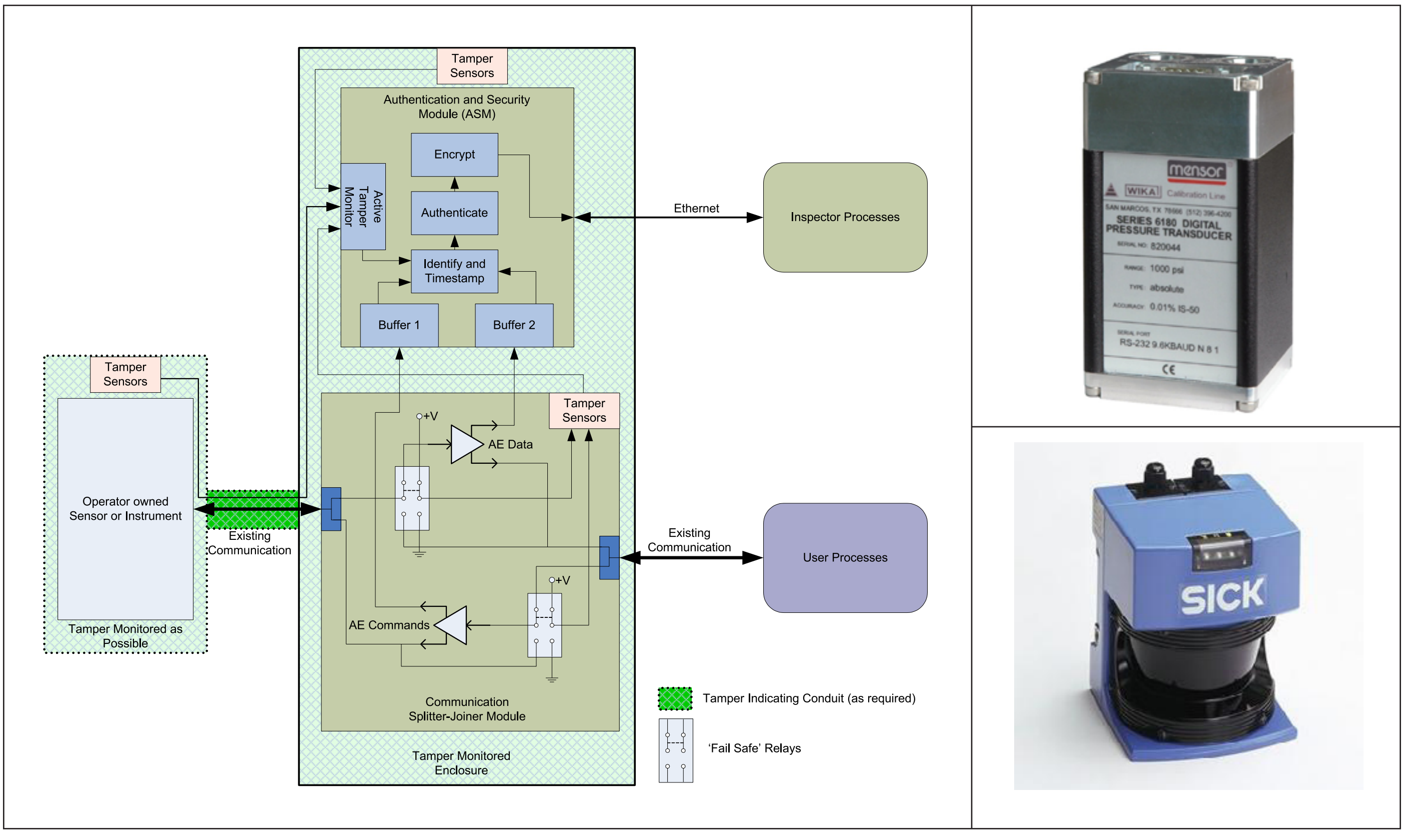

Figure: General Conceptual Architecture of the Enhanced Data Authentication System – EDAS, and photos of the two sensors used in the demonstration: pressure sensor and 3D laser based distance measurement system

After an initial setup, the EDAS prototype outputs an authenticated and encrypted data stream containing all the information generated by the sensor. The output data stream was then checked interactively with the original sensor-based data stream to verify the integrity of the data produced. Further, the EDAS prototype was also able to capture all the sensor configuration commands. These data was also replicated in the authenticated and encrypted EDAS output stream.

The discussion on the EDAS concept included concerns both from the inspector's perspective (accuracy, completeness, authenticity, confidentiality, meaningfulness) as well as from the operator point of view (non-interference and fail-safe operation). The team reported that thus far, it had demonstrated the ability of EDAS to meet inspector requirements. The next goal is to have a practical demonstration of the EDAS data sharing concept at a nuclear site under Safeguards while ensuring that operator requirements are fully met.

2.4. Interface between Safeguards and Security

The working group discussed the interface between Safeguards and Security in both meetings in 2010. Though discussions focused on the Containment and Surveillance perspective, there was a joint section with the NDA Working Group where the overall topic of the interface between Safeguards and Security was also discussed.

Indeed, though the application requirements may be considered different, there are many common points to Safeguards and Security, which should be investigated in view of potential synergies. Examples include equipment development, data security (including secure data transmission) concerns, access control, data review, etc. Specific aspects to each field should be identified to guarantee functional and procedural independence.

2.5. Requirements for C/S Systems under Integrated Safeguards

DG-Energy triggered the discussion on the impact of Integrated Safeguards in the European Union in existing Containment and Surveillance equipment. Because unannounced inspections create difficulties for EURATOM inspectors to participate and operator to prepare for these inspections, short notice random inspection schemes were agreed and implemented, if after the announcement time all operations are fully covered by surveillance. This approach somehow shifts the concept of Continuity of Knowledge. Surveillance data-streams, though permanent, are now randomly evaluated. Further, there is the need for larger local storage media due to longer and unplanned inspection intervals.

A solution to these difficulties relies on the increased use of Remote Transmission of Safeguards data between the plant and EURATOM headquarters at Luxembourg. An

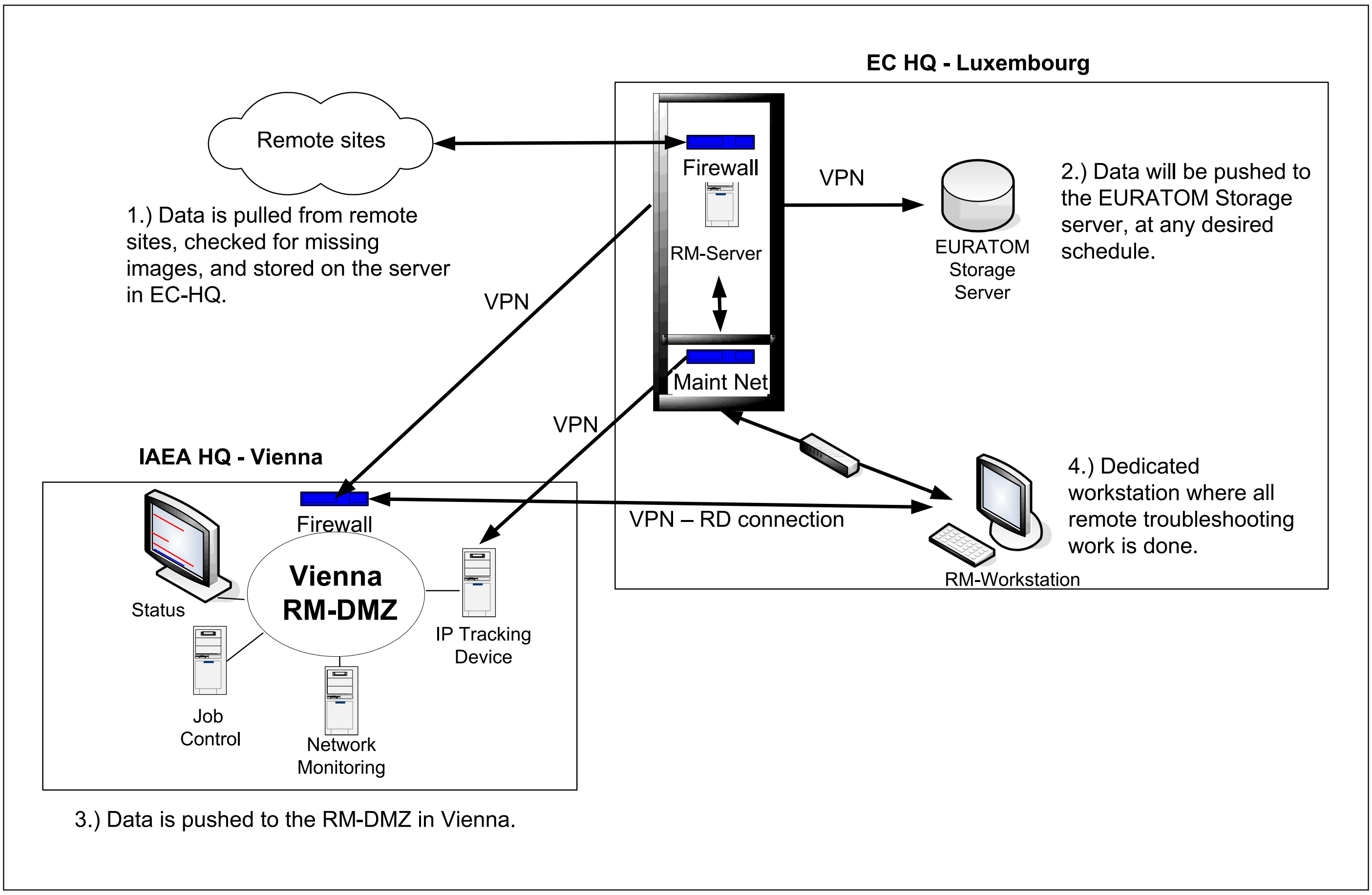

Figure: General Architecture of the Remote Data Transmission System agreed between DG-Energy and the IAEA

agreement on the sharing of remotely transmitted Safeguards data was reached between EC's DG-Energy and the IAEA. Apart the standard requirements on authentication, integrity and confidentiality, this agreement envisages a symmetric solution for data sharing, equipment maintenance and setup. The Remote data transmission concept is implemented stepwise with the agreement of EU member states. Steps include the transmission of (a) status of Health, (b) Safeguards data without surveillance images and, finally, (c) Safeguards images.

2.6. Visit to JRC SILAB: Seals And Identification Laboratory

The visit was devoted to the demonstration of current developments using commercially available RFID technology. In the first application, the RFID is used as the core component of a sealing system. Two systems are presented with low and high security features. In the second application, the RFID is used for a document managing system. In this application, a RFID tag is put on each page of the document. It is then possible to trigger an alarm whenever a document is retrieved from the cabinet and whenever a page is retrieved from the document.

3. C/S Compendium

The Working Group maintains a web based Compendium of all C/S instruments used by Safeguards inspectorates (typically, DG-ENERGY and IAEA). The computer platform supporting the Compendium will be changed to a more modern, wiki-based, platform. First tests have been successful and migration to the new platform is expected soon.

4. Future Activities

The working group is scheduled to meet twice per year. The following topics are scheduled to be discussed:

- C/S at Final Disposal Facilities (including Geological Repositories)
- Safeguards, Safety and Security: C/S perspective (continuation of the discussions)
- Remote system control
- Review of Surveillance Data Streams
- Technical Updates on new instruments and technologies

Report on the 2010 activities of the NDA Working Group Activities in 2010

Paolo Peerani and Anne-Laure Weber (former and new chairpersons)

The activity of the NDA-WG in 2010 was characterised by a strong interaction with other ESARDA working groups: two joint meetings were organised respectively with the DA-WG in May and with the C/S-WG in October. Moreover several members of the WG contributed significantly to the first meeting of the newly established NA/NT-WG held in Vienna in October.

Regarding the ongoing projects, an important step ahead was reached in the preparation of the document "Performance Values of NDA techniques for Waste Sentencing". The version 2 of the report was issued in September and can be considered a final draft. It is now undergoing a final revision round and the final version is expected to be published during 2011.

After the publication of the final report of the "ESARDA Multiplicity Benchmark" on the Bulletin issue no. 42 in November 2009, the group felt that further investigations would have been beneficial. Therefore a proposal for a deeper analysis of data has been elaborated by the WG. This proposal has been submitted to the participants of the benchmark and includes the study of:

- influence of detector parameters (poly density, gas pressure, active length,…)
- effects due to electronics (dead-time,…)
- uncertainty in sample model (geometry, material,…)
- nuclear data

A new topic that has raised a large interest among the WG members is the problem related to the shortage of He-3 and the research on valuable alternatives for its replacements in neutron detection. Several presentations have been made during the meetings on this subject, analysing the state-of-the-art and various R&D initiatives investigating alternative neutron detection technologies. As a spin-off of the discussions within the WG, a proposal for a project, called SCINTILLA, has been submitted for funding at the EC FP7 security call, where several WG members are involved.

In collaboration with the DA-WG, the working group has contributed to the finalisation of the IAEA document "International Target Values 2010". After a first draft issued by the IAEA in the second half of 2009, the comments and recommendations produced by the NDA-WG experts have been presented to IAEA in a Coordinated Expert Meeting held in Vienna on March 15^{th}-17^{th}. Following this meeting a revised version of the ITV tables was issued and presented to a joint NDA and DA meeting held in Luxembourg during the ESARDA Annual Meeting, where they were approved. The final version of the ITV-2010 document should be released early 2011 by the IAEA and will be published on the ESARDA Bulletin.

A joint meeting with the C/S-WG was held in Ispra in October with a two-fold objective:

- analyse the synergies between nuclear safeguards and security
- revise and update the common document "Guidelines on URMMS"

Four presentations from members of the two WG's have served as trigger to the discussions: the first from Baldwin and Funk that will produce a paper for the Bulletin, then Horvath enlarged the scope including also the third S (safety), Peerani elaborated on similarities and differences from a metrological perspective and finally Schwalbach presented an overview of monitoring techniques used by ENER in nuclear plants.

Finally the two WG's analysed the document on Unattended Remote Monitoring and Measurement Systems and found that, even though mostly valid, it would require some minor update and revision (in progress).

The ESARDA NDA-wg members provide support to the IWG-GST (Int.'l Working Group on Gamma Spectrometry Techniques). This working group, co-sponsored by ESARDA and INMM, gathers gamma spectrometry specialists both from Europe and America with the purpose to jointly work on problems related to isotopic measurements of U and Pu with special emphasis on code sustainability, standardisation and validation issues. ESARDA provides e-support by hosting and managing the website of the IWG-GST. Moreover it contributes to the development of a testing platform for gamma spectra evaluation codes. This platform will contain a collection of spectra that can be used by code developers to validate their new versions and by users to test and benchmark the performances of different codes. The architecture of the platform has been defined; the contents, structure and formats are described in a document that can be found on the website. Collection of spectra has started; currently approximately 200 spectra have been offered by ESARDA members and US laboratories as starting nucleus. A panel of experts is supposed to convene in 2011 to evaluate and select the spectra considered useful to be included in the platform. A comprehensive paper (Koskelo et al.: "Sustainability of Gamma-ray Isotopics Evaluation Codes") describing the platform and the list of available spectra has been presented at the 51st INMM Annual Meeting in July and is available in the conference proceedings.

The Working Group Training and Knowledge Management

Thomas Jonter, Montserrat Marín Ferrer and Sophie Grape

The main purpose of the work conducted by the working group Training and Knowledge Management (TKM) is to improve education and training in safeguards and nuclear non-proliferation for students and professionals in the European Union[1]. The TKM was created by ESARDA in 2004 with the aim to set up an annual course in safeguards and to develop a course syllabus to reduce the education deficit in the safeguards area. The purpose and aim go hand in hand with the goal to establish a European curriculum for Nuclear Engineering by European Nuclear Engineering Network (ENEN).

Since training in Safeguards and nuclear non-proliferation is in many respects strongly influenced by different traditions in different countries in the EU, the objective of the course is to create a common ground for the principles that are taught in line with international standards decided by the IAEA and the European Commission. Another objective is to incorporate the latest research and to include practical experiences by leading experts in the nuclear non-proliferation field. Hence the annually held course at the Joint Research Centre in Ispra, Italy, has the ambition to provide the participant with s a homogeneous and up-to-date course material in Safeguards and nuclear non-proliferation.

Description of tasks

An annual course has been conducted at JRC in Ispra since 2005, featuring a full five-days program with 1h lectures by experts in the field of nuclear safeguards. The program foresees every day a visit to one of JRC's safeguards laboratories and/or a classroom exercise. The course material, consisting of a complete set of presentations and literature, is be provided to the participants. Students can include this course, recognised by the Belgian Nuclear Higher Education Network (BNEN) and ENEN for 4ECTS, in their academic curriculum. To be quoted for this course, an additional Take-Home-Exam and a written paper must be completed.

This course is open to master degree students, in particular nuclear engineering students, but also to young professionals and international relations/ law students. It aims at complementing nuclear engineering studies by including nuclear safeguards in the academic curriculum. Based on the CVs of the participants the TKM tries to create synergies and interactions between technical and non-technical students in order to gain a more complete overview of different aspects within the fields of safeguards and non-proliferation represented in the course.

The basic aim of the course is to stimulate students´ interests in nuclear safeguards. The course addresses different aspects of the efforts to create a global nuclear nonproliferation system and how this system works in practice, such as e.g. the Treaty on Nonproliferation of Nuclear Weapons (NPT), safeguards technology, physical protection and export control. Also regional settings, such as the Euratom Treaty, are presented and discussed. The course content deals particularly with technical aspects and applications of safeguards i.e. how to implement the safeguards principles and methodologies within different nuclear facilities and how inspections are carried out by IAEA and Euroatom. Therefore, the course also presents an overview on inspections techniques, ranging from neutron/ gamma detectors, to design information verification, environmental sampling, etc.

Scope and content of the course

The basic information and subjects to be covered by the course/conference modules are:

- general background of legislation on safeguards relevant treaties and agreements in the EU and world-wide (survey, responsibilities of national and international institutions, situating nuclear safeguards in the overall Non Proliferation system)
- the nuclear fuel cycle: survey of technologies from mining to the final repository;
- basic principles of nuclear safeguards (accountancy and verification according to a structure of material balance areas, containment & surveillance, implementation of the safeguards principles in the nuclear fuel cycle)

[1] However, it should be mentioned that even non-EU states students have participates in the course. For example, around ten students from Russia have enrolled in the ESARDA course financed by the Swedish Radiation Safety Authority.

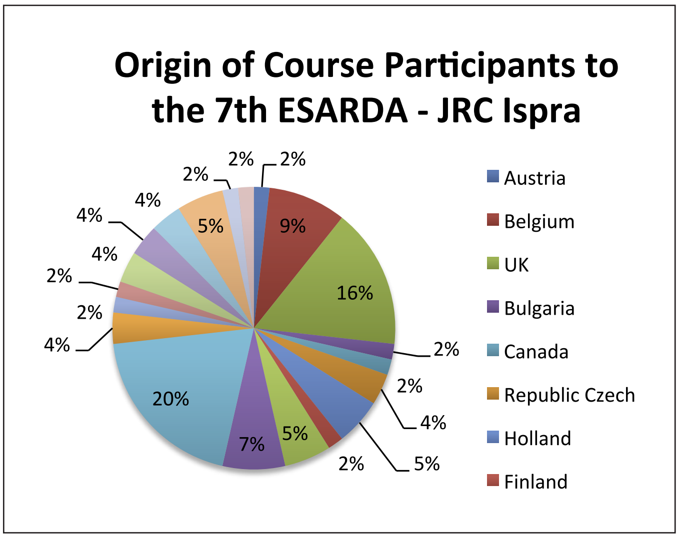

Figure 1: The 7th ESARDA course in Ispra. The course participants represented 15 different countries. The figure shows the number of participants from each of these countries.

- verification technologies (measurements and instrumentation): analytical techniques (destructive and non destructive assay), sealing, surveillance and monitoring techniques;
- evolution of safeguards approaches (strengthening of safeguards, integrated safeguards, Additional Protocol, information based safeguards).

Achievements

Since the first course was introduced in 2005, seven annual courses have been conducted with around 60 participants each year. A textbook, *Nuclear Safeguards and Non-Proliferation* (Ed. Greet Janssens-Maenhout), has been published containing articles developed and written by experts participating as lecturers in the courses. The publication of this book can be considered as a great success, but in the future it needs to be updated more frequently in order to incorporate the current knowledge and experiences according to the latest standards.

During 2011 two courses were held. The 7th ESARDA course in Ispra, March 28 – April 1, had 56 participants from 14 countries.

The second course – or the 8th ESARDA course – took place at Uppsala University in Sweden, September 12-16, 2011, and it was sponsored by the Swedish Radiation Safety Authority. The reason for having two ESARDA courses this year is due to the growing interest in among students and professionals to participate. In fact, today more than 100 applications are submitted to the course in Ispra and that means that around 40 % of all applicants have to be rejected.

At Uppsala University, 44 participants from 15 countries followed the one week course, comprising four days of lectures, one group exercise and one full day visit to SFR, the final repository for short-lived radioactive waste, located in Forsmark by the Baltic Sea about 100 kilometers north east of Uppsala. The excursion included an introductory presentation with a description of the facility, an underground visit in the facility and another presentation on nuclear power and the nuclear reactors in the Forsmark nuclear power station. A visit to one of the full-scale simulators of a control room in Forsmark was also part of the visit.

The course was highly appreciated by the students, due to its mixture of lectures on both a political and a technical level, which were given by very competent experts in the field. 23 professionals were involved in the course, either by giving lecturers and/or arranging with the organization of the course. 13 of them (57%) were affiliated with a Swedish institution (the Swedish Radiation Safety Authority, Uppsala University, Stockholm University and the Swedish company SKB). The remaining professionals represented the IAEA, the European Joint Research Centre (IRMM in Belgium), King's College London (UK), L'Institut de Radioprotection et de Sûreté Nucléaire (IRSN in France), the European Commission, the Ministry for Foreign Affairs (Finland) and the European Joint Research Centre (ITU in Germany).

Even though the course evaluation demonstrate that the courses have been highly valued by the participants, we believe that in the future we have to be able to better satisfy both participants (those who are more focused on academic knowledge and learning) and professionals (those who are more focused on practical work-related issues). One way to tackle this problem is to create parallel sessions, one for students and one for the professionals, within the same course.

Over the last years, the TKM has been very focused on initiating cooperation with other organizations dealing with training and education in the nuclear non-proliferation field.

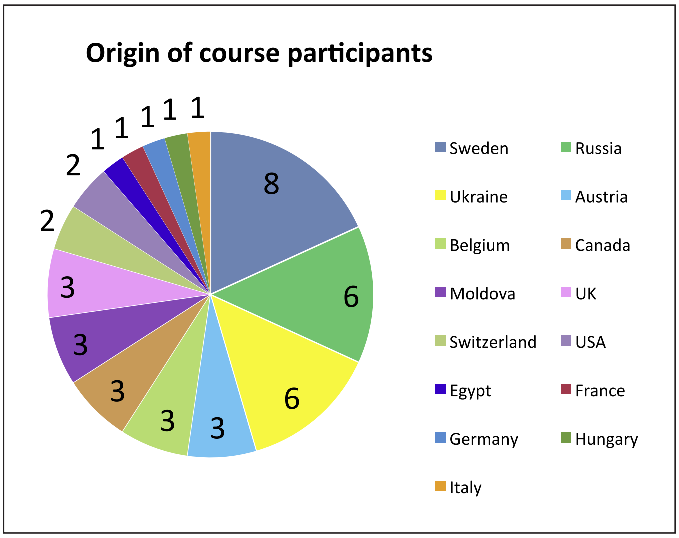

Figure 2: The ESARDA course in Uppsala. The course participants represented 15 different countries. The figure shows the number of participants from each of these countries.

Worth mentioning in this context, is the collaboration with INMM (The Institute for Nuclear Materials Management) in order to establish NuSaSET (Nuclear Safeguards and Security Education and Training), a joint ESARDA-INMM initiative to support nuclear Safeguards and Security Education and Training globally. A web-portal has been set up with the aim to provide support to professionals in the field of nuclear safeguards and nuclear security, especially to promote training and education of students (http://www.nusaset.org/)

The 9th Esarda Course on Nuclear Safeguards and Non proliferation will take place at the JRC Ispra (Italy) from 26th March to 30th March 2012.

9th ESARDA Course on Nuclear Safeguards and Non Proliferation

ESARDA Working Group on Training & Knowledge Management, M. Marin-Ferrer

1. Origin of the course

The knowledge retention problem in the nuclear field was acknowledged by the OECD in 2000. The United Nations study on disarmament and non-proliferation education (2002) made detailed recommendations for urgently required improvements. ESARDA, the European Safeguards Research and Development Association, reacted to these shortcomings with a strategy to tackle the problem and created a Working Group on Training and Knowledge Management (ESARDA WG TKM). The final objective of the ESARDA WG TKM is the setup of academic course modules to an internationally recognised reference standard.

This project is in line with the movement of establishing a European curriculum for Nuclear Engineering. Teaching in the Nuclear Safeguards field is indeed strongly influenced by national history. Thus the objective of the course is to provide a homogeneous material in Nuclear Safeguards and Non-Proliferation matters at the European and international level.

2. Learning objectives

This compact course is open to master degree students, in particular nuclear engineering students, but also to young professionals and international relations/ law students. It aims at complementing nuclear engineering studies by including nuclear safeguards in the academic curriculum.

The basic aim of the course is to stimulate students´ interests in safeguards. The course addresses aspects of the efforts to create a global nuclear nonproliferation system and how this system works in practice: the Treaty on Nonproliferation of Nuclear Weapons (NPT), safeguards technology, and export control. Regional settings such as the Euratom Treaty are also presented and discussed. In particular, the course deals with technical aspects and application of safeguards such as ways to implement the safeguards principles and methodology within different nuclear facilities. Therefore, the course will create an overview of inspection techniques ranging from neutron / gamma detectors to design information verification and environmental sampling, etc.

3. Course content

Introduction: The evolution of the Non Proliferation Treaty-regime; safeguards; international control regimes in theory and practice; and present trends in the nuclear nonproliferation efforts.

What is safeguarded: Definition of nuclear material that is subject to nuclear safeguards and related safeguards goals (significant quantity, timeliness and detection probabilities).

Where is it found: Description of the nuclear fuel cycle from mining to final repository, focusing on enrichment in the front-end and reprocessing in the back-end.

Which legal protection means exist: Overview on international and regional Non-Proliferation Treaties and established Institutions and Organisations.

What is the methodology to verify: Nuclear material accountancy principles and statistics for auditing.

How are inspections performed: Overview on inspector tools and their use to verify the nuclear activities as declared under the safeguards agreements (Non Destructive Assay, Monitoring, Containment/ Surveillance); additional safeguards measures under the Additional Protocol (complementary access, satellite imagery, environmental sampling) and how they are applied in field (storage facility, process facility, enrichment facility, research institute, spent fuel transfer).

How to control Import/ Export: Guidelines of the Nuclear Suppliers Group, trigger list and dual-use list. Means to combat illicit trafficking including nuclear forensics.

What additional information is offered: Collection of open source data and demonstration of some case studies (Iraq, 1993).

4. Practical organisation

The course features a full five-days programme with 1 hour lectures by a pool of experts in the field of nuclear safeguards. The programme foresees every day a visit to one of the JRC's safeguards laboratories and/or a classroom exercise.

The course material, consisting of a complete set of presentations and literature, will be provided to the participants. It is recommended that the students prepare themselves by means of reading the material on the website.

For this limited enrolment course early registration is recommended. A numerus clausus of 45 is introduced. The registration form can be found on:

http://esarda2.jrc.it/internal_activities/WC-MC/Web-Courses/index.html which should be completed and sent by the 15th January 2012 to JRC-NUSAF-SECRETARIAT@ec.europa.eu

University students can apply for accommodation free of charge, but only a limited number of places per university are available. Travel costs are not reimbursed by the JRC but there are no course fees and lunches are offered free of charge.

All participants are encouraged to write an essay on a given topic selected from a list, which is handed out at the end of the course. Up to two best essays can be selected for publication in the ESARDA Bulletin or for presentation in the poster session at the next ESARDA Symposium.

Students can include this course, recognised by BNEN/ENEN for 4ECTS, in their academic curriculum. To be quoted for this course an additional Take-Home-Exam is foreseen.

Participants are encouraged to register on the new **Nu**clear **Sa**feguards **S**ecurity **E**ducation and **T**raining web portal to be continuously informed about the organization of similar events: www.nusaset.org

Venue: JRC Ispra, Building 36, Amphitheatre

Schedule: From Monday 26th March at 8:30 to Friday 30th March 2012 at 17:00

Report on the Workshop on Direct Analysis of Solid Samples Using Laser Ablation-Inductively Coupled Plasma-Mass Spectrometry (LA-ICP-MS)

Organised by the ESARDA Working Group on Standards and Techniques for Destructive Analysis (WG DA)

Y. Aregbe[1], T. Prohaska[2], Z. Stefanka[3], É. Széles[4], A Hubert[5], S Boulyga[6]

1. European Commission-Joint Research Centre-Institute for Reference Materials and Measurements, Geel, Belgium
2. University of Natural Resources and Applied Life Sciences BOKU, Vienna, Austria
3. Hungarian Atomic Energy Authority
4. Hungarian Academy of Sciences, Institute of Isotopes, Budapest, Hungary
5. Commissariat à l'Énergie Atomique – CEA / DAM Ile de France
6. International Atomic Energy Agency – Safeguards Analytical Laboratories – IAEA-SAL, Seibersdorf, Austria

Abstract

The ESARDA Working Group on Standards and Techniques for Destructive Analysis (WG DA), in close collaboration with the Hungarian Atomic Energy Authority (HAEA) and the University of Natural Resources and Life Sciences (BOKU), organised a dedicated workshop on 'Direct Analysis of Solid Samples Using Laser Ablation-Inductively Coupled Plasma-Mass Spectrometry (LA-ICP-MS)'. The workshop was held in conjunction with the ESARDA Symposium, on 16 May 2011 at the Helia Conference Hotel in Budapest, Hungary. The workshop aimed to explore the potential of LA-ICP-MS for safeguards, non-proliferation, nuclear forensics and other applications. Safeguards authorities, fuel manufacturers, analytical laboratories and experts in the field of LA-ICP-MS were invited to participate in this workshop, to exchange views and information on the challenges and limitations of LA-ICP-MS in these areas. Forty representatives from the main European and international nuclear safeguards organisations, nuclear measurement laboratories, nuclear industry and manufacturers, and also experts from geochemistry and environmental sciences institutes, participated in this workshop. The plenary lecture was given by Dr. Joachim Koch from the ETH Zürich on 'Recent Trends and Advancements in LA-ICP-MS' followed by sessions focusing on the application of LA-ICP-MS in nuclear safeguards and nuclear forensics. The second session of this workshop was entirely dedicated to particle analysis with LA-ICP-MS and quality control. The findings and points of discussion from these sessions were further discussed in a working group using the 'World-Café' approach around four selected topics, ensuring that all workshop participants could benefit from the 'collective intelligence'. This report is a summary of the findings and points of discussion raised during the sessions and in the working group, including recommendations for research; instrumental development and data interpretation; reference materials and quality control; also emphasising different fields of application. As in previous workshops organised by the ESARDA WGDA, all participants recognised the need and the benefit of intensifying cooperation between the nuclear safeguards and nuclear forensics communities, nuclear industry and instrument manufacturers, and environmental sciences institutes. This report is an attempt to share the outcome of the workshop with a broader community.

Keywords: mass spectrometry; laser ablation; nuclear safeguards; nuclear forensics environmental sampling; earth sciences.

1. Introduction

The ESARDA WG DA seeks to emphasise the technical convergence of nuclear safeguards, nuclear forensics and nuclear security. One aspect to meet this objective is to strengthen the exchange beyond the safeguards community on dedicated technical topics relevant to all three fields using the WG DA as a platform. Previous workshops of the WG DA were dedicated to 'Measurements of minor isotopes in uranium in bulk and particle samples' [1] and to 'Measurements of impurities in uranium' [2].

One of the most powerful tools to detect undeclared nuclear activities is the analysis of environmental samples. Bulk analysis of collected swipe samples using Thermal Ionisation Mass Spectrometry (TIMS) or Multicollector Inductively Coupled Plasma-Mass Spectrometry (MC-ICP-MS) may be used, whilst methods for investigation of single particles can provide significantly more detailed information on the range of past activities in nuclear facilities. Nowadays, alongside other techniques such as Secondary Ion Mass Spectrometry (SIMS), ICP-MS combined with laser ablation (LA) sample introduction allows direct investigation of the isotopic composition of uranium and transuranium elements in single particles. In this technique, the material is ablated from a small area of a solid surface using a laser beam and swept to the ICP-MS by an argon or helium carrier gas. LA-ICP-MS has been successfully used for a wide range of different applications e.g. geological, forensics, materials sciences or nuclear applications. LA-ICP-MS (equipped with multi and single

Institution	Country
CAMECA	France
Centro Technologico da Marinha	Brazil
Commissariat à l'Énergie Atomique – CEA / DAM Ile de France	France
Commissariat à l'Énergie Atomique – CEA Marcoule	France
Department of Inorganic and Analytical Chemistry, University of Szeged	Hungary
European Commission – Directorate-General for Energy	European Commission
European Commission – Joint Research Centre-Institute for Transuranium Elements – EC-JRC-ITU	European Commission
European Commission – Joint Research Centre-Institute for Reference Materials and Measurements – EC-JRC-IRMM	European Commission
Geological Institute of Hungary	Hungary
Hungarian Atomic Energy Authority	Hungary
Institute of Isotopes, Hungarian Academy of Sciences	Hungary
International Atomic Energy Agency – Safeguards Analytical Laboratories – IAEA-SAL	United Nations
Korea Atomic Energy Research Institute – KAERI	South Korea
Laboratory of Inorganic Chemistry / ETH Zurich	Switzerland
National Nuclear Laboratory	United Kingdom
Nuclear Research Center Negev	Israel
Paul Scherrer Institut – PSI	Switzerland
Sellafield Site	United Kingdom
Thermo Fisher Scientific (Bremen)	Germany
University of Leicester and NERC Isotope Geoscience Laboratory	United Kingdom
University of Natural Resources and Applied Life Sciences BOKU	Austria

Table 1: List of participating institutions

collector detectors) is a promising new technique for nuclear safeguards and forensics applications. However, there are still several questions on this field, which have to be answered for routine application of the method. During the annual working group meeting in 2010, the WG DA members expressed the need to address the topic of direct analysis of solid samples with LA-ICP-MS in a dedicated workshop, exchanging opinions and knowledge with invited experts in this field. The aim was to explore and enhance knowledge on applications outside nuclear safeguards in order to improve the level of knowledge on potential and limitations of this technique. The announcement was distributed to all the WG DA members and posted on the ESARDA web-site. The interest in this workshop was overwhelming, and the number of participants (about 40!!) exceeded by far the expectations of the organisers, recalling that this workshop was dedicated to a single specific instrumental technique. Representatives from the main European and international nuclear safeguards organisations, nuclear measurement laboratories, nuclear industry and manufacturers, and also experts from geochemistry and environmental sciences institutes participated in this workshop. This positive response to the announcement was a confirmation that the WG DA had chosen a topic that was of great interest to a broad community.

The institutions that participated in the workshop are listed in Table 1.

2. Objectives of the workshop

The main workshop objective was to explore the potential of LA-ICP-MS via knowledge exchange between experts from nuclear safeguards, forensics, industry and environmental sciences and to draft recommendations in respect to:

- Needs and requirements of Environmental Sample (ES) analysis for IAEA Safeguards
- Fundamental research
    - Measurement performance
    - Limits of detection and quantification
- Instrumental development and limitations

- Fields of application
  - bulk and particle analysis,
  - forensic analysis and nuclear trace analysis for safeguards
  - needs in other areas (fuel samples,…)
- Internal and external quality control

3. Workshop structure

Zsolt Stefanka from the HAEA welcomed the participants on behalf of the hosting organisation. Yetunde Aregbe, in her capacity as chair of the WG DA, opened the workshop with a short review on the recommendation from the ESARDA WGDA 'Report on the Workshop on Measurements of Minor Isotopes in Uranium' that new methods, like LA-ICP-MS, should be investigated further and mature applications should be developed [1]. During that workshop, in 2008, the following points were identified for further research and development to enhance the potential of LA-ICP-MS applications:

- Detector linearity, transient signals
- Interferences, detector dark noise
- Uranium hydrides correction
- Composition and Matrix-matched standards
- Energy filter adjustments
- Femto-second lasers
- Simultaneous measurements of U, Th, Pu

Subsequently, the workshop objective, structure and practicalities were outlined to the participants. Nine presentations from workshop participants were given in one plenary and two topical sessions. The findings and points of discussion from these sessions were further discussed in a working group using the 'World-Café' approach around four selected topics, ensuring that all workshop participants could benefit from the 'collective intelligence'. Subsequently, the outcome of the discussions around the four selected topics was presented to all workshop participants and first recommendations were drafted. The workshop was closed by Yetunde Aregbe, acknowledging the HAEA for hosting the event and the contribution and good cooperation between the experts from nuclear safeguards, forensics, industry and environmental sciences.

3.1. Plenary session

The plenary session addressed the technical background and recent developments in LAICP-MS. The plenary lecture, entitled 'Recent trends and advancements in LA-ICP-MS', was given by Joachim Koch, ETH Zürich (CH).

Laser ablation was introduced as a solid sampling technique in the early years of ICP-MS and quickly developed into a mature and widely accepted technique [3, 4]. The method itself is based on a laser ablation system, which acts as sample introduction device, and an inductively coupled plasma mass spectrometer, which acts as element/isotope specific detector. A laser beam (diameters range typically between 2 and 500 µm) is focused onto the surface. The laser beam, directed onto a solid surface within a dedicated laser ablation cell, leads to the evaporation and removal of material from a solid surface resulting in, ideally, round-shaped craters. The most commonly used lasers are Nd/YAG lasers, with a fundamental wavelength of 1064 nm. The frequency is quadrupled to generate UV laser light. The most common laser wavelengths are 266, 213 and 193 nm. Although 266 and 213 nm Nd:YAG lasers are the most commonly used systems, 193 nm lasers show better ablation properties with respect to equal ablation rates in samples with different absorption behaviour and similar matrix. Moreover, 266 nm lasers produce larger particles compared to 193 nm lasers (in the order: particle size produced by 193 nm lasers < 213 nm lasers < 266 nm lasers [5]). 193 nm lasers are usually based on ArF excimer lasers which are more complex and expensive compared to solid state laser systems. Originally, lasers used pulse lengths in the nanosecond (ns) time range: nowadays, femtosecond (fs) lasers are increasingly popular even though they are significantly more expensive compared to commercially available nanosecond laser systems. The ablated material is transported via an Ar or He gas stream to the inductively-coupled plasma, which acts as ionisation source resulting in the formation of preferably single charged monoatomic ions. The ions are further separated in the mass spectrometer according to their mass/charge ratio and finally detected with a single or with multiple collectors. The latter are applied when isotope ratios with the highest precision are required. In general, all isotopes of the periodic table of elements can be analysed by these devices. Although significant progress has been made since the early days of LA-ICP-MS, fundamentals of the technique are still being explored.

J. Koch addressed in the plenary lecture the general aspects of ICP-MS including the most common mass analysers (quadrupole, magnetic sector field and time of flight). Moreover, the recently presented MC-ICP-MS using a Mattauch Herzog Geometry was discussed, which allows the simultaneous acquisition of isotopes along the entire mass range. This is seen to be an asset in laser ablation measurements where only short transient signals are available (i.e. single particle measurements) [6]. Special emphasis was given to recent advances, particularly to the introduction of femtosecond laser ablation as a sample introduction system for ICP-MS [7]. The particle size distribution of the latter systems lead to <5% of particles larger than 0.1 µm. Special interest was directed to the visualisation of laser ablation in order to gain more information about the flow dynamics within an ablation cell and the modelling of the flow using computational fluid dynamics (CFD).

3.1.1. Application in nuclear safeguards and nuclear forensics

The first session after the plenary was dedicated to LA-ICP-MS for nuclear safeguards and nuclear forensics applications, chaired by Amelie Hubert from the Commissariat à l'Énergie Atomique CEA/DAM. In this session, selected examples of the use of LA-ICP-MS for nuclear safeguards and forensics were given by experts from ITU, MTA IKI and the IAEA.

The advantages of LA-ICP-MS, such as minimal sample preparation; local analysis; precise isotope ratio measurements; and rapidity, were compared against the disadvantages, such as interferences; calibration; heterogeneity; lower sensitivity; and some geometric and instrumental limitations. For nuclear forensic analysis, the advantages using LA-ICP-MS are that almost any material can be analysed without chemical pre-treatment, and that a short analysis time is needed to determine the isotopic and elemental composition of illicit material, using only a few mg of the material, thus minimising waste. Studies have been carried out at ITU to compare LA-MC-ICP-MS with a traditional destructive analysis approach for the determination of age of nuclear materials via the $^{230}Th/^{234}U$ 'clock'. As a result, the detection limit of ^{230}Th was found to be at 33.6 pg g-1, which is approximately 100 times higher than for a full destructive analytical method. In spite of relatively high uncertainty, the "age" of a uranium material can be obtained quite fast, and is within the larger uncertainties useful for nuclear forensics applications, making LA-ICP-MS a valuable method for screening of material. LA-ICP-MS is also applicable to measurement of the isotopic composition of uranium oxide particles, preferably not smaller than 10mm, where the combined relative uncertainty measurement results for the major isotope range between 1 – 5%.

The Institute of Isotopes of the Hungarian Academy of Sciences (MTA IKI) is a technical support organisation of the Hungarian Atomic Energy Authority. It has been assigned by the government to attribute found or seized nuclear material of unknown origin. ICP-MS as a destructive method and LA-ICP-MS as a quasi non-destructive analytical technique are applied at the MTA IKI to perform measurements on such materials. Some special methods have been developed for characterisation of nuclear material using LA-ICP-MS, e.g. age dating by $^{230}Th/^{234}U$, determination of the enrichment from U isotope ratio measurements and determination of the reprocessing properties of nuclear material [8]. A method for the analysis of single particles originating from safeguards swipe samples has been developed. Particles are collected from the surface of the swipe samples using the vacuum impaction method. Subsequently they are studied using SEM and analysed by the LA-ICP-MS technique. Simple and rapid analysis of single particles, down to a size of about 10 µm, is possible using a laser ablation system with low laser energy [9].

Detection of undeclared nuclear activities relies on the collection of high quality samples, sophisticated, accurate and appropriate analytical techniques and the interpretation of analytical results using a variety of information and data evaluation methods. The IAEA is currently considering the potential use of LA-ICP-MS, to expand the "Safeguards Toolbox" through the identification and implementation of novel detection and monitoring techniques for the detection of undeclared nuclear activities, materials and facilities. The range of potential applications in the IAEA analytical services includes impurity analysis, isotope analysis, age determination and particle analysis. As mentioned by other experts during the workshop, the IAEA acknowledges the merits of LA-ICP-MS in view of spatial resolution, direct analysis of small samples, short analysis time and low detection limits, and is investigating its limitations and routes for further development. For impurity bulk analysis, LA-ICP-MS does not seem to be well suited, and is probably more useful for impurity distribution analysis (nuggets etc.) [10] Also, for isotope bulk analysis of U samples, limitations concerning peak tailing, interferences and matrix effects result in lower precision and accuracy. However, LA-ICP-MS might be useful for rapid screening, or for measurements of highly radioactive samples [11, 12]. The advantage of LA-ICP-MS for age determination is in the direct analysis of small samples and particles of 10100mm. Currently, the age limit with this technique at the IAEA is about 10 -15 years for uranium, due to observed peak tailing, interferences and element fractionation (Th-U fractionation) [13].The performance of the LA technique might be further improved by using modern MC-ICP-MS and femtosecond lasers. Limitations of LA-ICP-MS for particle analysis result from the relatively large spot size and the difficulty to localise a single particle that is smaller than 1µm [14]. The IAEA is convinced that ongoing R&D in instrument development and towards higher sensitivity will further increase the potential for application of LA-ICP-MS in safeguards-related analyses.

3.1.2 Particle Analysis

Session 2 focused on particle analysis and was chaired by Sergei Boulyga from the IAEA Safeguards Analytical Services (IAEA-SGAS). Presentations were given by experts from the British Geological Survey, BOKU-WIEN, Nuclear Research Center Negev and CEA/DAM.

Results from the uranium isotope analysis of (depleted) UO_2 particles by LA-MC-ICP-MS were presented by the University of Leicester. In the 1960s, many residents at the National Lead Inc. (NL) site in Albany, New York were exposed during years of U emissions. The aim of the study was to reconstruct the history of emissions, and maybe to identify and traced forensically the feedstocks of depleted uranium. Such a long time of residence of U oxide particles in soil and environment allows study of degradation and dissolution from aerosol deposition. Sand, soil and dust samples from the contaminated area were collected, screened and analysed with different instrumental techniques: ICP-Quadrupole Mass Spectrometry (QMS) for concentrations and isotope ratios; XRF for metal concentrations; MC-ICP-MS for high precision isotope ratios; LA-MC-ICP-MS for individual particles; Scanning Electron Microscope/Energy Dispersive X-Ray Spectroscopy (SEM-EDX) for particle elemental screening and Transmission Electron Microscopy (TEM) / X-Ray Diffraction (XRD) / X-Ray Absorption Spectroscopy (µXAS) for particle mineralogy. The depleted uranium oxide particles originated from the NLI plant and were recovered from dry household dusts. These depleted uranium oxide particles are respirable. The particles, mounted on epoxy and polished for imaging prior to analysis, were ablated with a new wave 193nm solid state laser at its lowest power setting, with a laser spot ~15-20µm in diameter. For isotopic analysis, an Axiom MC-ICP-MS with mixed ion counter and Faraday detectors was used. The ^{236}U and ^{234}U ratios were measured on the IC, the others on Faraday detectors. ^{235}U was measured on both for cross calibration. For quality control, natural uraninite mineral grains were repeatedly measured. It was concluded that the precision of the measurement results is clearly sufficient for source attribution (Paducah gaseous diffusion plant) and nuclear forensic purposes, but improvements need to be done when measuring particles < 5mm. Using a high sensitivity instrument, such as the Neptune or the Neptune Plus, in combination with integration of a single laser pulse, could enable measurement of major and minor uranium isotopes even on small particles with LA-ICP-MS. There is definitely a need for interlaboratory comparison samples, to investigate the limits of LA-ICP-MS towards small particle analysis.

Thomas Prohaska, from the University of Natural Resources and Life Sciences, Vienna, presented the state of the art work using LA-MC-ICP-MS for ^{234}U, ^{235}U, ^{236}U to ^{238}U ratios of single particles based on earlier work on determining the isotopic composition of uranium and fission products in radioactive environmental microsamples [15]. He presented the application of nanosecond laser ablation (UP 193, ESI–NWR Division, Electro Scientific Industries, Inc., Portland, CA, USA) to an MC-ICP-MS (Nu Plasma HR, Nu Instruments Limited, Wrexham, UK) for the direct analysis of U isotope ratios in single, 10–20 µm-sized, and U-doped glass particles. The use of a deceleration filter showed clearly an improvement of the abundance sensitivity by a factor of 10. A total combined uncertainty budget was presented, including the contributions from repeatability along with the hydride rate, the ion counter yield, blank, dead time and peak tailing. The work demonstrated clearly the applicability, reliability and robustness of LA-MC-ICP-MS for the direct analysis of individual particles with respect to their U isotopic composition, with total combined uncertainties of less than 10% relative standard uncertainty.

E Elish from the NRCN presented results from cooperation with the JRC-ITU on the “Application of Laser Ablation-ICP-MS for Forensic Analysis and Nuclear Trace Analysis for Safeguards”. In this study, the same samples were measured at NRCN, by LA-MC-ICP-MS and LA-ICP-QMS, and at ITU, by SIMS. Original swipe samples from JRC were used for the analysis: UO_2 particles, UO_2 particles + Al_2O_3 Fine Powder and UO_2 particles + Al_2O_3 Particles. Pre-tracking of particles by SEM was investigated via fixing the particles, taking the coordinates, and relocating the particles with a grid. The 3 grids coordinates were determined using the SEM, 3 locations on each grid were determined, and each particle’s coordinates (X, Y) were determined by SEM. The same coordinates of the 3 grids are re-determined using the LA software with respect to one of the locations to find each particle’s coordinates. Ablation was then done at or around the calculated particle coordinates. Without SEM pre-tracking, the JRC particles were ablated in a single point, in a raster of several points, and along points on a (segmented) line, and it was attempted to locate them by the Laser Optics, which is of course inferior to SEM pre-tracking. With this approach only large particles can be detected. On the other hand, SEM pre-tracking is very time consuming. In any case, improvements are still needed for both approaches to determine the limits of detection for particle analysis for safeguards application. In another study carried out at NRCN, it was shown that detection of occupational exposure to uranium from LA-ICP-MS analysis of a single hair strand is possible. The limit of detection with this technique is 19ng U g^{-1} hair [16].

F Pointurier from the CEA presented results from the analysis of uranium micro-particles using ns-LA-ICP-QMS at CEA. These results were also compared with results from other methods such as SIMS and Fission Track–TIMS (FT-TIMS). Samples measured were IAEA swipe samples and the NUSIMEP-6 certified test sample [17]. The first step in localising the particles is to immobilise the particles by embedding them in a collodion layer on a polycarbonate disk. For direct location, SEM equipped with software for automated detection of uranium particles is used. This is a relatively rapid technique, but it is difficult to locate particles <1mm. This has been improved at the CEA with the Field Effect Generator technology, providing higher resolution to detect more and smaller particles using the Gun Shot Residue (GSR) software. For indirect location of particles, the fission track technique is used. Each polycarbonate disc is covered with a solid state nuclear track detector disc. Both discs are welded and corresponding marks are made on each of them. Irradiation takes place in Orphée (Saclay, France) for about 60s at a total flux of 10^{15} thermal neutrons · cm^{-2}, leaving tracks originating from the small portion of ^{235}U that undergoes fission [18]. After chemical etching, both polycarbonate disks are positioned on the optical microscope stage. The marks recorded before irradiation help to identify the location of the particles that gave the respective fission tracks. The isotopic composition of the particles is measured with a laser ablation system coupled to an ICP-QMS. The ablation rate is 20 Hz, the spot size is 100 µm, and the diameter of the ablated area is about 100 µm. Particles are ablated in a single point of 100 shots with a delay between 2 shots of 100 ms. The total duration of the ablation is 10s. Particles can be relocated within a range of 20-50 µm and are analysed by ICP-QMS. To test the performance of this system, NUSIMEP-6 uranium reference particles with a size <1µm were measured to compare with previous NUSIMEP-6 results by FT-TIMS and SIMS. In addition, real-life swipe samples previously analysed with FT-TIMS were measured. As a conclusion of these tests it was observed that, combined with fission tracks, ns-LA-ICP-QMS compares favourably with TIMS at CEA for isotopic measurements. It is faster and more sensitive for very small particles < 1 µm, but combined with SEM/GSR, ns-LA-ICP-QMS shows lower precision compared to SIMS. In any case, ICP-QMS is not ideal for minor isotopes and has significant limitations in precision and accuracy for very noisy signals: therefore, MC-ICP-MS would be the instrumental ICP technique of choice together with optimised laser ablation parameters. Overall, ns-LA-ICP-MS is definitely an alternative technique for isotopic analysis of micrometer-size U particles, provided particles can be located beforehand and fixed. The success of LA-ICP-MS for safeguards applications also depends on improvements in particle location.

3.1.3. Quality control

The last presentation was given by IRMM and dedicated to the ongoing Nuclear Signatures Interlaboratory Measurement Evaluation Program NUSIMEP-7 on *Uranium isotope amount ratios in uranium particles.* NUSIMEP-7 is the second IRMM interlaboratory comparison on uranium particle analysis organised in support of the European Safeguards (DG ENERGY), the International Atomic Energy Agency's (IAEA's) Network of Analytical Laboratories (NWAL) and laboratories in the field of particle analysis. Since the previous interlaboratory comparison NUSIMEP-6, IRMM succeeded in modifying the preparation of uranium oxyfluoride particles via the hydrolysis of well-certified UF_6 in the gas phase by improving their quality to better simulate real-life uranium particles found on swipe samples. The efforts to optimise the preparation of the NUSIMEP-7 samples were verified by SEM at IRMM and by SIMS at ITU and the IAEA-SGAS. This optimised method was used to prepare new certified test samples for NUSIMEP-7 with single and with double isotopic enrichment. Measurement of the sample with two different isotopic particle depositions will be challenging for participants using LA-ICP-MS, because the NUSIMEP-7 particles are of size < 1mm. The final NUSIMEP-7 participant report will be available in due time via the IRMM web-site [19].

3.2. Working Group

After the sessions with presentations from invited speakers, the second part of the workshop was dedicated to discussing the findings from those presentations in a working group and to draft a set of recommendations on the applicability of LA-ICP-MS in safeguards, forensics and trace analysis, including technical advancements in ablation and detection; on data evaluation; and on methodologies for identification of relevant signatures.

Due to the number of workshop participants, and the limited time frame of only one workshop day, the ESARDA WG DA tried for the first time a different approach for the working group discussion. The aim of the working group was to review in more detail the state of the art, the fields of application, the limitations and strategies for improvements of LA-ICP-MS. Therefore the working group discussion on 'Potential and limitations of LA-ICP-MS for nuclear safeguards and trace analysis' was organised in a so-called "World-Café" [20], chaired by T Prohaska from BOKU-WIEN with assistance from Y Aregbe, E Széles and Z Stefánka. The 'World-Café' is a workshop method based on the assumption of a collective knowledge. The participants are guided to interact in a constructive way in their discussions, where each participant can express his/her point of view. They are spread within different topics, where they deal with a specific question. To each of the topics a facilitator is assigned. After a set time, the participants change within the topics, get a résumé by the facilitator of the topic and restart the discussion with the next question related to this topic.

Within the ESARDA workshop, the participants were divided in four groups, of about eight participants per group, to discuss the four topics in line with the objective of the workshop as listed below.

1) Fundamental research and method development
2) Instrumental development
3) Fields of application
4) Quality control

Around these four topics, the following questions were raised in four rounds and discussed by the four working groups in a rotational sequence:

- State of the art (What do we have?)
- Wish list (What do we really want?)
- Limitations (What does not work?)
- Recommendations (What should we do?)

Discussions between experts from the various fields of application proved to be highly beneficial. Each workshop participant contributed to each of the four topics, therefore all participants could benefit from the 'collective intelligence' in the room. This approach enabled a first set of recommendations per topic to be drafted and presented by the four workgroup hosts at the end of the workshop day. The following four paragraphs are an elaborated version of that set of recommendations, identified by the workshop participants.

3.2.1. Recommendations for fundamental research and developments

This topic dealt with measurement performance of LA-ICP-MS and limits of detection and quantification. From the discussions, it was evident that fundamental research is the prerequisite for accurate data assessment and that a significant amount of basic and fundamental research in the field has to be conducted. It is evident that the method has high potential, but still, little is understood about the fundamental processes in the analytical setting. Basically, proper investigation of mass fractionation effects has to be accomplished. This is seen as prerequisite for accurate isotope ratio determination. In addition, interferences – also with strong matrix dependence – have to be studied accordingly. E.g. hydride formation can become a crucial topic, especially if external calibration by combining liquid sample introduction with laser ablation is accomplished. For basic research purposes, U-rich material was recommended as basis material. To obtain reliable data from transient signals, appropriate data evaluation strategies have to be considered and evaluated along with laser ablation strategies (e.g. single pulse ablation). As a further development in LA-ICPMS, it was considered of high importance to make use of the multielement capabilities of the ICP-MS, not only considering the parallel measurement of different isotopic systems (e.g. Pu along with U) but also the simultaneous detection of impurities in single particles. Nonetheless, still, proper sampling strategies are missing when particle sizes of less than 10µm are under investigation. Particle analysis by LA-ICP-MS is related to a number of analytical challenges, such as: (i) difficulty of particle localisation; (ii) the necessity to detect very small amounts of isotopes; (iii) complex matrices that result in specific interferences and matrix effects; (iv) treatment of very short transient signals. A better understanding of processes that govern the interaction of laser irradiation with solid particles, aerosol formation (in particular, size distribution of aerosol particles), the temporal characteristics of aerosols atomisation and ionization in the ICP, as well as the processes of separation and detection of ions in the mass analyser, is of crucial importance for the development of efficient LA-ICP-MS protocols for isotope analysis of actinides in particles. It is recommended to review the available information and to investigate further the following issues with the focus on laser ablation of uranium particles:

- Dependence of size distribution of the LA-generated aerosol and fractionation effects on the applied laser ablation parameters (type of laser ablation system, wavelength, pulse length, irradiation power density etc.).
- Behaviour of LA-generated aerosol in the ICP-MS, including atomisation efficiency of uranium as well as spatial and temporal distribution of produced ions in the ICP. Effect of ICP parameters on isotopic mass fractionation and on interference rate
- Extraction efficiency of ions in the interface, efficiency of ion transport in the mass analyser, mass fractionation effects in the mass analyser.
- The mechanisms of detection and evaluation of transient signals
- Matrix effects, such as effect of sample matrix on mass fractionation and creation of specific interferences. In particular, consider potential approaches for (simultaneous) assessment of the chemical composition of sample matrices by using other analytical methods in parallel with isotope ratio measurement by LA-ICP-MS

A major concern was raised towards the proper establishment of uncertainty budgets for the analysis of samples with LA-ICP-MS, as most of the relevant contributors are still either not understood or simply neglected. This will be taken up in the next ESARDA WG DA workshop dedicated to 'Uncertainties in Nuclear Measurements'.

3.2.2. Recommendations for instrumental development

This topic dealt with instrumental development and limitations of LA-ICP-MS. The conclusions were a perfect follow-up of the recommendations for further fundamental research (see 3.2.1.). It was agreed by all participants that the current ability of LA-ICP-MS for particle localisation is not very satisfactory. Therefore, a major part of the discussion was dedicated to the detection of small particles where an improvement of localisation strategies was recommended. The use of a scanning mode of laser ablation for particle localisation is limited, due to the fact that it would be quite time consuming when scanning with high resolution over a large surface area and LA is a destructive method and the scanning process would destroy the object of interest. In addition, the use of a relatively large spot size (a few hundreds micrometers) is required for a time-efficient scanning, which would lead to a mix up of individual particles. The use of a sufficiently small spot size would significantly extend scanning, so that several days would be required for scanning of a single sample. So, the combination of other, non-destructive, methods for particle localisation techniques with LA-ICP-MS was considered beneficial. The use of SEM with adequate coordination or interferometry was addressed. Moreover, the idea of using fission track mounts directly with LA-ICP-MS was seen as an asset. A general optimisation of laser ablation cells for this particular task is also required. The development work should focus on improving stage control to allow more precise and reproducible control of particle positions and to relocate particles accordingly (also when combining, for example, with SEM). Furthermore, optimisation of cell geometry to reduce memory effects and to increase transmission rate of aerosol into the ICP is needed. Instrumental sensitivity is still considered a limitation to single particle analysis. Even though sensitivity has improved significantly within recent years, it is still an issue, especially when investigating minor isotopes. This goes along with developments of the detectors. A special challenge is the further development of adequate multidetector arrays as they are used in ICP-MS instruments with Mattauch Herzog Geometry. Improvement in the interface design was an important issue, improving the sensitivity and/or reducing potential interferences. Recent developments of, for example, the jet interface already points in this direction. Although the mass range is not an issue in Mattauch Herzog based geometries, where the whole mass spectra can be imaged, a limited mass range is still an issue in MC-ICP-MS using Nier Johnson geometry. Extended mass ranges are still considered as a significant asset in these machines, along with the use of a significantly larger number of secondary electron multipliers. The laser ablation system was considered as a source for improvement, as well. Small spot sizes along with homogeneous beam profiles are needed. The development of fs laser ablation goes in this direction, but with the major problem that these instruments are bulky, expensive and demanding in their operations. In addition, laser ablation cells have room of improvement, especially if considering combination with other techniques (e.g. SEM, FT) or an automated ablation strategy. Summarising, the workshop participants expressed strong recommendations towards instrumental development, to (re)locate particles <10 µm, to increase sensitivity and abundance sensitivity as well as development of robust detectors and procedures for data evaluation that are appropriate for handling short ion packages, which are produced by ablation of uranium particles.

3.2.3. Recommendations for fields of application

This topic dealt with application of LA-ICP-MS for forensic analysis and nuclear trace analysis (bulk and particle) for safeguards and the needs in other areas. Even though the fields of application seemed to be innumerable, the following core applications were identified:

For comprehensive investigation of nuclear safeguards and nuclear forensics related samples, complex analytical methods are needed. Typically, the available sample amount is limited, therefore requiring combined analytical techniques to carry out measurements simultaneously on the same sample (e.g. single particle, nuclear fuel pellet). The combination of a number of techniques for particle analysis is seen as a significant benefit for accessing the inherent information in such samples. Combination of Laser Induced Breakdown Spectroscopy (LIBS) and time of flight mass spectrometry (TOF-MS) were identified as possible approaches. Major and minor isotopes of U and Pu are measured for safeguards and nuclear forensics purposes. It is recommended to consider the potential of LA-ICP-MS for determination U and Pu isotopes in particles with complex matrices that are difficult to analyse by other methods. Furthermore, it is recommended to investigate further the ability of modern LA-ICP-MS instruments for the determination of minor isotopes of uranium and for age determination of uranium particles. A developed method for Pu age dating was seen as quite challenging, but could be used as an important independent investigation method for the production date of nuclear fuel material.

For nuclear forensics, a very important task is the analysis of the “non-nuclear part” of the confiscated samples (e.g. packaging materials). Elsewhere, combination with other isotopic systems can yield additional information concerning the provenance of the samples. For example, a precise Pb or Sr isotope ratio analysis could provide information indirectly regarding the geographical origin of the nuclear or other radioactive materials. The simultaneous determination of isotopic systems other than U and Pu are a particularly significant challenge if it comes to small particles (see 3.2.2), of a size less than 1 µm, but this would be needed to enable the analysis of typical safeguards swipe samples with LA-ICP-MS: an area where still little knowledge exists.

3.2.4. Recommendations for quality control

This topic dealt with the availability of quality control tools and the needs of “tailor-made” internal and external specific for LA-ICP-MS. The experts all agreed on the need for a variety of particle standards and interlaboratory comparison schemes. Furthermore, mixed standards of particles with interfering elements and particles mixed with dust are needed. There was common agreement that NUSIMEP-6 and NUSIMEP7 are very useful to the community, but that, particularly for LA-ICPMS, the particle size should be larger. There was a unanimous need expressed for more Interlaboratory Comparison schemes. The main recommendations for further development of quality control tools specific for LA-ICP-MS are listed below.

- Recommendation for further development of particles standards:
  - mixed isotopic and elemental composition
  - particle size variation (0.5 – 10µm)
  - possible matrix/interfering elements (Al, Pb,...)
  - non-particle standards for instrumental calibration
  - U, Pu particles mixed with dust
  - Particles on transparent substrate
- Recommendation for particles required as QC for age dating:
  - Size 1 – 10µm
  - varying enrichment

In addition it was recommended to make biomarker standards available for trace analysis (e.g. hair form population exposed to higher concentrations of specific trace elements due to geographically related natural environmental conditions). Reference materials for speciation might also be useful. In particular, uncertainty estimations were seen as crucial and once more the question came up whether it would make sense to adopt a similar concept to the International Target Values for fissile and nuclear material analysis in environmental sample analysis [21]. The next ESARDA WGDA workshop on 'Uncertainties in Nuclear Measurements' will discuss these topics in more detail.

4. Summary and Outlook

The ESARDA WGDA adopted recently 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 [22]. Part of meeting this objective is the organisation of dedicated workshops on advancements and applicability of analytical techniques to read signatures in nuclear material and environmental samples, with participation beyond the safeguards community, bringing together experts from safeguards, nuclear forensics, earth sciences, and particularly from industry. This was the first WGDA workshop fully dedicated to a single instrumental technique with a clearly defined technical focus. The discussions held in the working groups and in the plenary meeting resulted in broad recommendations. The different measurement communities participating in the workshop agreed that LA-ICP-MS has proven its strengths in geological, forensics, materials sciences and has a potential for nuclear applications, particularly for particle analysis. At the same time it is clear that there are still some technical challenges before LA-ICP-MS can become a routine method in safeguards. Advances in research and development, also from the side of instrument manufacturers, would benefit all user communities.

Issues that need to be addressed:

- Technical advancements in ablation – femto-second lasers
- Localisation and analysis of particles <10 µm on swipe samples
- Improvement in sensitivity and abundance sensitivity for determination of minor isotopes
- Interferences, Uranium hydrides correction
- Detector calibration, application of energy filters, transient signals
- Interferences and element fractionation for simultaneous measurements of U, Th, Pu
- Limits of detection for different application needs (nuclear safeguards, nuclear forensics, trace analysis)
- Availability of well-certified particle reference materials (size >1 µm)
- Availability of matrix-matched 'real-life' bulk and particle reference materials
- Organisation of interlaboratory comparisons
- Uncertainty estimation has to be carried out according to the *Guide to the expression of uncertainty in measurement (GUM)* [23]
- LA-ICP-MS as stand-alone and/or complementary method – timelines, automisation

Overall, the outcome of the workshop exceeded by far the expectations of the organisers with respect to participation, discussions and to meeting the objectives. The present report is a further attempt of the WGDA to share the outcome of technical discussions and findings with a broader community using ESARDA as platform. The next dedicated workshop on '*Uncertainties in Nuclear Measurements*', organised by the ESARDA WGDA in close collaboration with the International Atomic Energy Agency Safeguards Analytical Services (IAEA-SGAS), will be held at the IAEA-SGAS Seibersdorf Laboratories, Austria, from 8-9 November 2011. The focus of this workshop is an exchange between reference measurement institutes, safeguards laboratories, nuclear and environmental material analysts and, in particular, operators, investigating major contributions to the final measurement uncertainties that depend upon the material and technique applied. The workshop will be open to ESARDA WG DA members and a limited number of participants from expert and research institutes [24]. As a result of the workshop, we expect a clear picture from state-of-the-art to state-of-practice along with recommendations on approaches in uncertainty estimation, on consistency of measurements carried out by nuclear laboratories and by operators with the GUM approach (ITV2010) [21, 23].

5. Acknowledgements

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6. List of Acronyms

- **ES** – Environmental Sampling

- **FT-TIMS** – Fission Track Thermal Ionisation Mass Spectrometry
- **ICP-MS** – Inductively Coupled Plasma Mass Spectrometry
- **LA-MC-ICP-MS** – Laser Ablation Multi Collector Inductively Coupled Plasma Mass Spectrometry
- **LA-ICP-QMS** – Laser Ablation Inductively Coupled Plasma Quadrupole Mass Spectrometry
- **LIBS** Laser Induced Breakdown Spectroscopy
- **(LG)-SIMS** – (Large Geometry) Secondary Ion Mass Spectrometry
- **NWAL** – Network of Analytical Laboratories
- **RM** – Reference Material
- **SEM/EDX** – Scanning Electron Microscope/Energy Dispersive X-Ray Spectroscopy
- **SIMS** – Secondary Ion Mass Spectrometry
- **TEM** Transmission Electron Microscopy
- **TIMS** – Thermal Ionisation Mass Spectrometry
- **TOF-MS** time of flight mass spectrometry
- **WG DA** – Working Group on Standards and Techniques for Destructive Analysis
- **XAS** X-Ray Absorption Spectroscopy
- **XRD** X-Ray Diffraction

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Technical sheet

COMPUCEA for on-site accountancy verification

1. Objective of the technique

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 analytical results are quickly and directly reported to the inspectors on-site, eliminating the necessity to ship samples to a Safeguards laboratory and thus ensuring timeliness. Against this is the requirement to dissolve solid samples, involving the use of operator facilities including weighing systems, and the additional inspector time required at the facility.

2. Presentation of the technique

2.1. 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 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.

2.2. Measurement techniques involved

Uranium concentration determination by L-edge densitometry

The technique is based on X-ray absorption edge spectrometry with a miniaturised X-ray source and a high-resolution Peltier-cooled silicon drift detector for uranium elemental assay. The schematic setup is shown in Fig. 1 (left side). 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: an example is shown in Fig 1 (right side). The characteristic jump of the photon transmission at the L-absorption edges of uranium can be seen. 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 evalua-

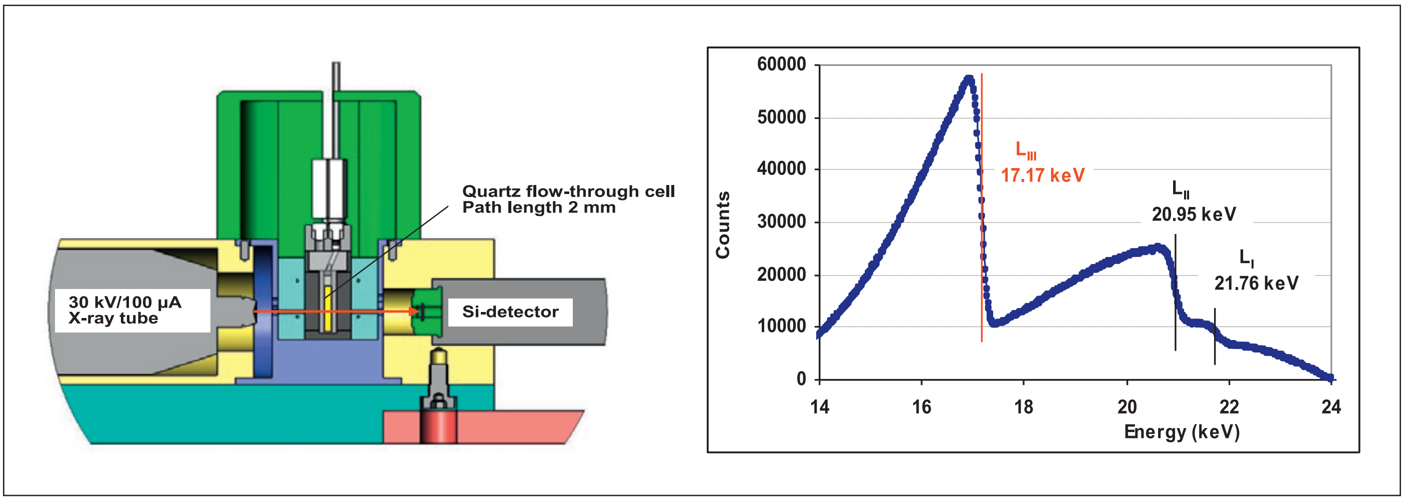

Figure 1: L-edge densitometry with an X-ray continuum used in COMPUCEA 2nd generation. Left: Measurement setup. Right: Sample spectrum.

tion 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 principle of gamma spectrometry for uranium isotopic determination is explained in more detail in [4]. The ^{235}U enrichment measurement in the 2nd generation of COMPUCEA (Fig.2) is based on the counting of the ^{235}U 186 keV gammas of a defined amount of uranium in solution in a well-defined counting geometry. The detector used is a

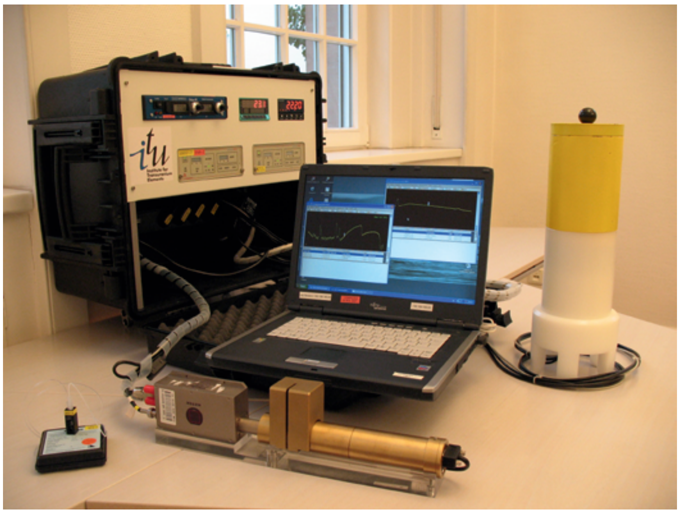

Figure 2: Setup of COMPUCEA 2nd generation equipment (L-edge densitometer in the front, $LaBr_3$(Ce) detector with shielding on the right) with electronics and computer for experiment control and data acquisition.

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 $LaBr_3$ (Fig.2) compared to High-Performance 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 [5] 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 by a Monte Carlo simulation.

2.3. International Target Values for measurement uncertainty

In the field of International Safeguards for nuclear materials, International Target Values (ITVs) for measurement uncertainties have been established for all relevant measurement techniques [6]. The corresponding ITV2010 for COMPUCEA are listed in Table 1, assuming a counting time of 1000 seconds. The actual uncertainties observed during infield campaigns are well within these ITVs.

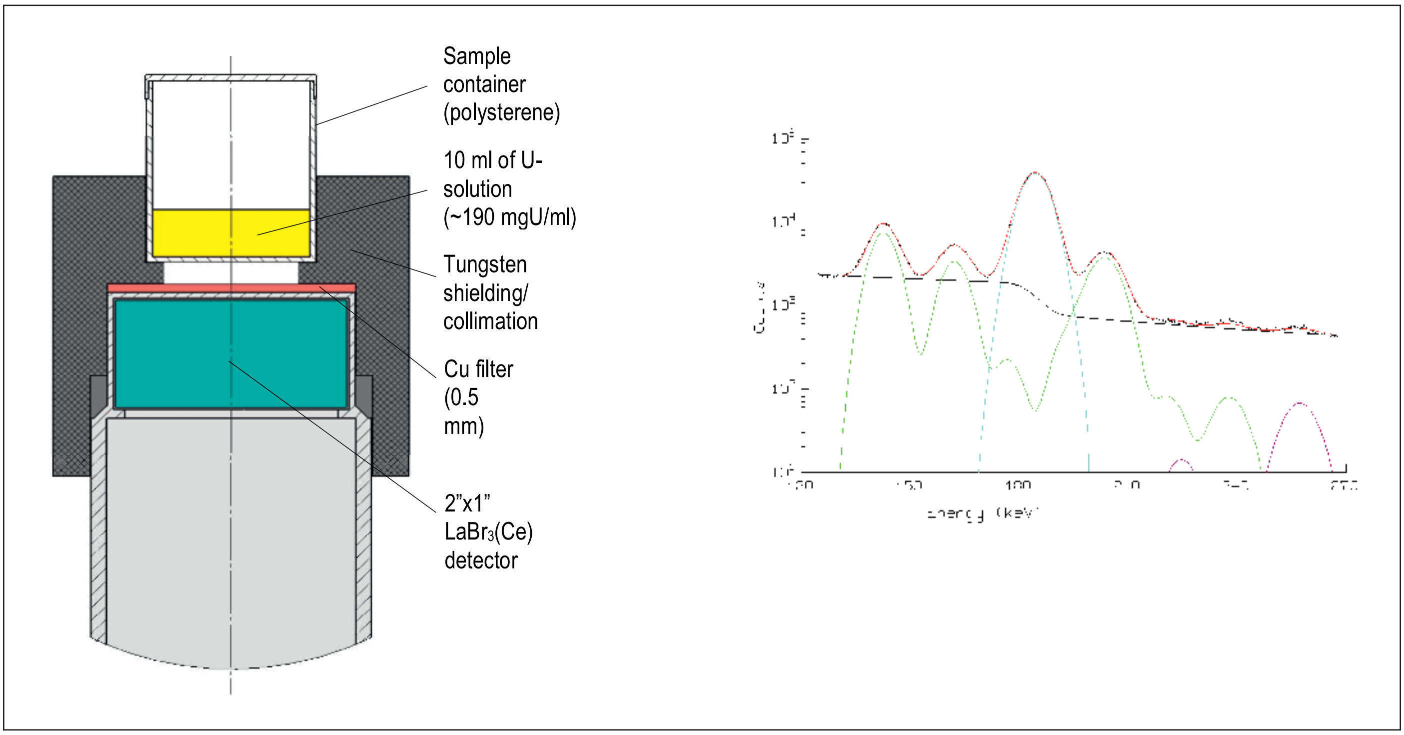

Figure 3: Setup for gamma spectrometry with a $LaBr_3$(Ce) detector (left) and fitted gamma spectrum obtained from a LEU sample (right).

Analysis	Combined relative Uncertainty (%)
U-concentration	0.28 a)
^{235}U abundance	0.45 a)

a) For a counting time of 1000 s.

Table 1: International Target Values 2010 for COMPUCEA.

3. Additional information and useful links – References

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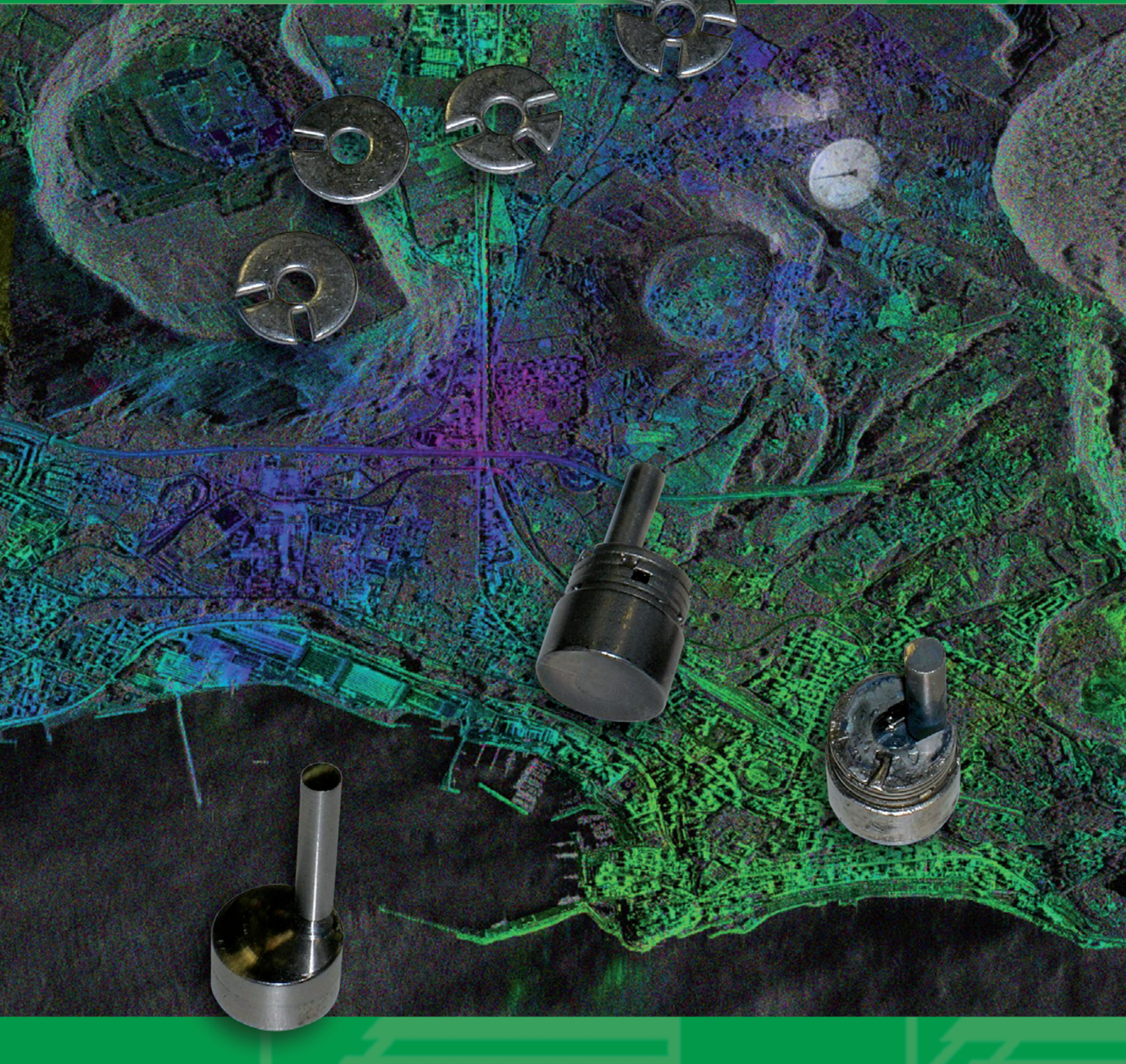

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