Standardization of JRC developed Nuclear Mass Spectrometry Methods with ASTM-International

Drafting ASTM Standard for the "Double Spike" (DS/TIMS) Test Method by JRC-G.2/METRO

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Abstract

In 2017 the JRC-G.2/METRO team has contributed to standardization of methods developed at JRC for nuclear mass spectrometry in various ways.

Firstly, in early 2017 the revision of the ASTM standard document for the "classical" total evaporation (TE) method, written by K. Mathew (LANL, US-DOE) and S. Richter (JRC-G.2/METRO), was published with designation ASTM C1672-17. The document contains a detailed description of the TE method for U and Pu measurements, which was originally developed at JRC in the late 1980s. S. Richter from JRC-G.2/METRO contributed to the revision of ASTM 1672 in particular to the newly defined requirements for calibration and quality control for TE measurements, as well as by providing new algorithms for uranium and plutonium concentration measurements by IDMS (Isotope Dilution Mass Spectrometry).

Secondly, during the year 2017 a new draft standard document has been written by S. Richter (JRC-G.2/METRO). The document contains a detailed description of the "Double Spike" method (DS/TIMS) for high precision $^{235}\text{U}/^{238}\text{U}$ isotope ratio measurements. The first draft has been submitted for the C26-05 sub-committee ballot on 04/09/2017. The double spike method for TIMS was used successfully for the validation program of the Cristallini method for sampling of UF$_6$, to which staff from the unit JRC-G.2 in Geel/Belgium and the SGAS/IAEA contributed successfully.

Thirdly, S Richter (JRC-G.2/METRO) revised the draft "New Practice for Sampling and Preparation of UF$_6$ using the ABACC-Cristallini Method", which was drafted by P. Mason (NBL Program Office, US-DOE) and O. Pereira (IPEN, ABACC) and which will include the validation report as an annex. This support from JRC-G.2/METRO takes place within the frame of the new "Action Sheet 43: Collaboration on Certified Reference Material Development and Safeguards Measurements Quality Assurance" signed between JRC and US-DOE in 2017, and the collaboration agreement between ABACC and the JRC signed in 1999.
1 Introduction

ASTM International, formerly known as the American Society for Testing and Materials (ASTM), is a globally recognized organization for the development of international voluntary consensus standards. The ASTM Committee C26 on the Nuclear Fuel Cycle was formed in 1969. C26 standards play a significant role in all important aspects to the nuclear fuel cycle. C26 meets twice a year, usually in January and June, with about 50 members attending over one week of technical meetings. C26 has 10 technical subcommittees. One of them, C26.05, is dealing with "Methods of Test", which includes standardized methods for the analysis of nuclear material for the nuclear fuel cycle. This also applies to mass spectrometric methods as there are used and further developed at JRC-G.2 and for which JRC-G.2 provides the material standards (i.e. reference materials) for calibration. The subcommittee C26.05 has implemented several small task groups, e.g. for U and for Pu methods, environmental methods and spectroscopy.

During the recent years, ASTM International has changed from being an American-only organization into an internationally recognized standardization body. In particular, for the standardization of materials and methods for the nuclear fuel cycle used by industry (like URENCO, AREVA) and safeguards organizations (like the IAEA), ASTM International has become the main partner for standardization. In order to strengthen the international character of its standardization efforts, the ASTM C26 committee started organizing some of its sub-committee meetings outside the American continent, e.g. in 2013 a meeting was held in cooperation with CEA/CETAMA in Aix-en-Provence, France, and in 2016 a meeting took place in Vienna, Austria, in collaboration with the IAEA. This latest meeting was combined with a workshop on "Analytical Developments, Reference Materials and Statistical Applications in the Nuclear Fuel Cycle", which included presentations by several staff member of unit JRC-G.2.

The collaboration between JRC-G.2 and ASTM-International was initiated in 2013 due to the invitation of S. Richter (JRC-G.2) by M. v. Wijnkoop (URENCO) to develop an ASTM standard document for the so-called "modified total evaporation" (MTE) method for the determination of uranium isotope abundances by thermal ionization mass spectrometry (TIMS) [1]. Since several years this method is known to provide a significant improvement for measuring in particular the "minor" isotope ratios 234U/238U and 236U/238U for certification work at JRC-G.2 as well as for routine nuclear safeguards measurements at the IAEA, ORNL and JRC-G.II.6.


During the year 2017, the revision of another important standard document for nuclear mass spectrometry, the document ASTM C1672 for the so-called "classical" total evaporation was published. This method was first developed at JRC [2] and subsequently further described by IAEA (International Atomic Energy Agency) [3] and LANL [4] (Los Alamos National Laboratory, United States). It has become the standard method for analysing the isotopic composition and isotope amount concentrations of uranium and plutonium samples for nuclear safeguards purposes around the world. Therefore, the revision of this standard document ASTM 1672 was considered very important. In order to guarantee accuracy of measurements for nuclear accounting and safeguards purposes, it has to be ensured that these measurements are performed in a reliable and trustworthy way. The first page of ASTM C1672 is shown in Annex-1.

The ASTM standard document for the TE method can be obtained through ASTM (https://www.astm.org/Standards/C1672.htm). The standardization of the TE method...
was also highlighted in the JRC news: https://ec.europa.eu/jrc/en/science-update/newly-revised-standard-astm-c1672-17-based-jrc-methodology

Most recently, within 2017 S. Richter (JRC-G.2) has prepared a new draft standard document for the so-called "Double Spike" method (DS/TIMS) for high precision $^{235}\text{U}/^{238}\text{U}$ isotope ratio measurements [5, 6]. The first draft has been submitted for the C26-05 sub-committee ballot on 04/09/2017. The double spike method for TIMS was used successfully for the validation program of the Cristallini method for sampling of UF$_6$, to which staff from the unit JRC-G.2 in Geel/Belgium and the SGAS/IAEA contributed successfully [7].

Within ASTM-international an international group of experts from LANL, JRC, IAEA and the nuclear industry are working together in order to provide revised and improved standard documents for frequently used methods, in order to assist the users for performing their analyses in a way that can be relied upon and defended to the safeguards and governmental authorities. The contribution of JRC-G.2 is considered very valuable in this context.

2 Experimental

The "Double Spike" method (DS/TIMS) for high precision $^{235}\text{U}/^{238}\text{U}$ isotope ratio measurements has been used for many years already, because it provides a remarkably better precision by a factor of about 5-10 compared to other commonly used TIMS methods like the "classical" total evaporation (TE) [2-4] or "Modified Total Evaporation" (MTE) [1]. This is due to the fact that the certified $^{233}\text{U}/^{236}\text{U}$ ratio of the double spike, e.g. IRMM-3636a, with a ratio $^{235}\text{U}/^{238}\text{U}=1.01906(16)$ certified on 01/07/2007 [5], can be used for an internal mass fractionation correction of the measured $^{235}\text{U}/^{238}\text{U}$ ratio throughout the entire measurement duration of a sample loaded on a filament. Due to the improved precision the double spike technique is already being used widely for scientific applications, in particular in geochemistry and cosmochemistry, for investigating the age and source of natural uranium samples, geochemical or physical processes [8-9], or for determining important half-life values of long-lived isotopes (i.e. $^{234}\text{U}$ and $^{230}\text{Th}$) for age determinations [10]. Based on the experience with the double spike method, JRC-G.2 proposed to ABACC and IAEA to use this method as a tool for the investigation of the Cristallini sampling process for UF$_6$ with the best possible precision and accuracy for the $^{235}\text{U}/^{238}\text{U}$ isotope ratio. For the so-called "minor" ratios $^{234}\text{U}/^{238}\text{U}$ and $^{236}\text{U}/^{238}\text{U}$ the MTE method was selected as the best choice at SGAS/IAEA [7].

3 Results

The work on the new draft standard document for the "Double Spike" method (DS/TIMS) was proposed and agreed upon during the ASTM-International meeting of subcommittee C26.05, "Methods of Test", on January 31$^{st}$, 2017. (S Richter could not attend the meeting due to other commitments at JRC-G.2). The relevant parts of the meeting minutes are shown below.
Opening of the C26-05-03 Task Group Meeting:

Most of the subsequent discussions were not relevant within the context of this report and therefore not shown here. The meeting was closed at 1:41pm and re-opened (after lunch) at 2:35pm:

Closed the spectroscopy Group 1:41
Reopened 2:35 – By Mike Brisson

Re: Email from Stephan Richter – proposed new standard – TIMS Double Spike Standardization, task Group including Non-C26.05 members Rebecca Thomas expressed an interest in joining the Task Group.

“Dear Mike,

Very good. I have got confirmation from Sebastien Mialle also that he will join the TIMS/DS standardization group. I also asked Lars Borg from LLNL, because he is using a 233U/236U Double Spike as well, but he didn’t pick up the method and spreadsheet from me (like Sebastien and Joe from the IAEA), so he could be a bit more independent as a contributor and reviewer, I hope his funding will be approved. So this is the task group composition:

Stephan Richter, JRC-G.2
Kattathu Mathew, LANL
Joseph Hiess, IAEA
Sergei Boulyga, IAEA
Sebastien Mialle, JRC-II 6
Lars Borg, LLNL (to be confirmed)

I wish you a successful January meeting and I hope to be able to join you next time again.
Regards,
Stephan
Dr. Stephan RICHTER”

Reclosed 2:39

After the meeting, a "collaboration area" was set up on the ASTM website by S Richter, and a task group formed, the description of the collaboration area is shown below.
Within the following months, the draft was written by S Richter (JRC-G.2) and reviewed by K. Mathew (LANL), S. Mialle (JRC-G.II.6) and J. Hiess (IAEA). The final draft was submitted to the C26-05 sub-committee ballot on 04/09/2017.
At the end of the balloting period, all comments will be retrieved and any questions discussed by email and/or at the next upcoming ASTM C26-05 subcommittee meeting, scheduled for January 21st-24th, 2018, in New Orleans. After that, a new revision will be prepared and submitted to the next following concurrent sub- and main committee ballot in spring 2018. Depending on the approval, the final document can be released in summer 2018.

Besides the drafting of the standard document for the double spike method, S Richter also revised the draft "New Practice for Sampling and Preparation of UF₆ using the ABACC-Cristallini Method", which is authored by P. Mason (NBL Program Office, US-DOE) and O. Pereira (IPEN, ABACC). The draft was discussed at the ASTM C26-05 meeting in June 2017 in W Conshohocken (PA). (S Richter could not attend the meeting due to other commitments at JRC-G.2). The relevant parts of the meeting minutes are shown below:

Most of the discussion is not relevant within the context of this report and therefore not shown here. Under the agenda point "new standards", the following comments were made regarding the Cristallini sampling method for UF₆:
Subsequently, S Richter also joined the subcommittee C26-02. The work on this document is still ongoing, and it is also planned to include a revised version of the validation report presented at the 58th Annual INMM Meeting 2017 [11] as an annex in order to give evidence for the trustworthiness of this new sampling method to all future users. It is foreseen to submit a draft version to a subcommittee ballot prior to the next upcoming ASTM C26-02 subcommittee meeting, scheduled for January 21st -24th, 2018, in New Orleans.

4 Conclusions

As in previous years, JRC-G.2 contributed to the standardization of methods for nuclear mass spectrometry which are relevant and often used in nuclear safeguards measurements worldwide. JRC-G.2 is currently involved in the preparation of two important new documents to be released in 2018.

It is worth mentioning that within November 2017, a visit of the ASTM President K. Morgan, to be accompanied by the Vice President G. Grove and the European representative of ASMT, Ms S. Gobbi, to the JRC-G.2 Geel site is planned. This will be combined with a meeting between the ASTM President K. Morgan and M. Betti, the Head of Directorate G for Nuclear Safety and Security of the JRC.

The role of standards is high on the policy agenda with the European Commission [12]. The JRC collaborates with international bodies and organisations in the US and across the globe to harmonise scientific techniques and standardise analytical processes. After the publication of the ASTM standard document ASTM 1832-16 for the JRC-developed "Modified Total evaporation" (MTE) technique in 2016, and the ASTM standard document ASTM 1672-17 for the "classical" total evaporation (TE) method in 2017, the draft standard for the double spike method (DS/TIMS) and the contribution to the Cristallini sampling method for UF$_6$ are further examples of JRC's input on standardisation in the nuclear field. The expertise developed at JRC-G.2 on a frequently used standard mass spectrometry technique will lead to the implementation of a significantly improved standard applicable in nuclear safeguards and nuclear industry world-wide.
References


List of abbreviations and definitions

ASTM American Society for Testing and Materials, United States
DS Double Spike
DOE Department of Energy, United States
IAEA International Atomic Energy Agency, United Nations
IRMM Institute for Reference Materials and Measurements (formerly). The nuclear activities of IRMM are continued at JRC - Unit G.2, named "Standards for Nuclear Safety, Security and Safeguards" (SN3S)
JRC Joint Research Centre, European Commission
LANL Los Alamos National Laboratory, Department of Energy, United States
METRO Metrological tools for nuclear Safeguards, Security & Safety
MTE Modified Total Evaporation method, used in TIMS (see below)
NBL New Brunswick Laboratory (formerly), now renamed as "NBL Program Office"
SGAS Safeguards Analytical Services (IAEA), located in Seibersdorf, Austria
SRNL Savannah River National Laboratory, Department of Energy, United States
TE Total evaporation method, used in TIMS (see below)
TIMS Thermal Ionization Mass Spectrometry

List of figures
No figures

List of tables
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Annexes

1. First page of the ASTM standard document C1672-17. The complete document can be obtained through ASTM, but cannot be posted completely here: (https://www.astm.org/Standards/C1672.htm).
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