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Evaluating the status of uranium isotope ratio measurements using an inter-laboratory comparison campaign

S. Richter*, A. Alonso, J. Truyens, H. Kühn, A. Verbruggen, R. Wellum

Institute for Reference Materials and Measurements (IRMM-JRC-EU), Retieseweg 111, 2440 Geel, Belgium

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Abstract

The REIMEP 18 (Regular European Inter-laboratory Measurement Evaluation Programme) campaign for the measurement isotopic ratios of uranium in nitric acid solution was completed in December 2006. The task for all participating laboratories was to measure the uranium isotopic composition of four uranium samples ranging from depleted to slightly enriched uranium. With 71 participating laboratories REIMEP 18 has become the largest nuclear isotopic measurement campaign organized by IRMM so far. Participation in this kind of measurement campaign is an integral part of the external quality control required for nuclear safeguards laboratories worldwide. For the first time also a significant number of academic laboratories, mainly from the geochemistry area was included.

Certification measurements were carried out at IRMM using state-of-the-art mass spectrometric methodology. A MAT511 UF_6-gas source mass spectrometer (GSMS) was used to determine the $n(^{235}U)/n(^{238}U)$ ratios and a TRITON thermal-ionization mass-spectrometer (TIMS) for the minor isotope ratios $n(^{234}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$. Verification measurements on ampouled samples were performed successfully prior to sample shipping and showed good agreement with the certified ratios.

The results of the REIMEP 18 campaign confirm in general the excellent capability of nuclear safeguards and scientific laboratories in measuring isotopic abundances of uranium, although some problems were discovered for the measurements of the minor isotope ratios $n(^{234}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$ and the calculation of measurement uncertainties for isotope ratios in general. This paper describes the outcome of the REIMEP 18 campaign. It includes a graphical evaluation and discussion of the results, an evaluation of the applied measurement and calibration techniques and a discussion of conclusions and actions to be taken.

Keywords: Isotope mass spectrometry; Uranium; Inter-laboratory measurement campaign; TIMS; ICPMS

1. Introduction

Measurements to determine the isotopic composition of uranium samples are done in a variety of industrial and scientific areas. First, because of the industrial use of uranium as fuel in nuclear power reactors, the relative isotopic enrichment of the fissile uranium isotope ^{235}U has to be quantified within each section of the nuclear fuel cycle, e.g., the enrichment facilities, the power plant operating facility, the reprocessing plant and finally the waste handling facility. The isotopic composition of uranium material going through the nuclear fuel cycle is subject to careful verification analyses by national as well as international nuclear safeguards authorities such as the IAEA and EURATOM inspectorates. Additionally, in order to prevent the use of uranium for undeclared or clandestine purposes, analyses of uranium samples acquired during possibly un-announced inspections at suspected nuclear sites are performed by the IAEA and EURATOM, etc. Second, the uranium isotopic composition is also measured in many scientific disciplines, such as geochemistry for disequilibrium studies or geochronology.

Uranium has four long-lived, naturally occurring isotopes, ^{234}U, ^{235}U, ^{236}U and ^{238}U. The presence of ^{236}U at the very low level of about 10^{-11} relative isotopic abundance in uranium ore has been confirmed by accelerator mass spectrometry (AMS) [1–3]. Additionally the non-naturally occurring isotope ^{233}U plays an important role as analytical tool, because it is an ideal spike material for isotope dilution analyses. It is also

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* Corresponding author. Tel.: +32 14 571701x652; fax: +32 14 571863.
E-mail address: stephan.richter@ec.europa.eu (S. Richter).

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produced in small amounts in various nuclear fuel cycles, for instance by decay of ^{237}Np.

Due to their relatively higher abundances the isotopes ^{235}U and ^{238}U are often called the major isotopes, whereas the isotopes ^{234}U and ^{236}U are often called the minor isotopes. The $n(^{235}\mathrm{U})/n(^{238}\mathrm{U})$ isotope ratios to be measured in the laboratory cover a quite large range. Natural uranium is characterized by $n(^{235}\mathrm{U})/n(^{238}\mathrm{U})$ isotope ratios of about 0.00725 with a variability of about 0.05% [4,5]. Higher $n(^{235}\mathrm{U})/n(^{238}\mathrm{U})$ isotope ratios – so-called enriched uranium – are the consequence of industrial isotopic enrichment processes which are needed for uranium to be used in nuclear power reactors (ca. 3–5% enrichment) or even nuclear weapons (>90%). Lower values – so-called depleted uranium – are generated as the by-product of the enrichment processes.

Measurements of the so-called minor ratio $n(^{234}\mathrm{U})/n(^{238}\mathrm{U})$ are performed for various reasons. They first serve as an additional tool to indicate the origin of nuclear samples or various types of isotope enrichment processes. Additionally the $n(^{234}\mathrm{U})/n(^{238}\mathrm{U})$ ratio is used in disequilibrium studies for geochemical research. The minor ratio $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ is also of great interest and the values to be measured cover a large range from about 10^{-11} for natural samples up to ca. 10^{-2} for enriched material. Any measurement of a $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ ratio significantly different from natural uranium indicates a nuclear reaction, e.g., neutron capture of ^{235}U, possibly caused by anthropogenic influence, which makes the measurement of the $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ ratio an important nuclear safeguards tool.

Due to the scientific and political or legal relevance of nuclear isotopic measurements all safeguards laboratories need a reliable quality management system to ensure their measured values are acceptable. Nuclear analytical laboratories are required to demonstrate their measurement capability on a regular timely basis. One way of demonstrating measurement capability is to participate in interlaboratory comparisons. For this reason, IRMM has organized quality control campaigns for measurements of uranium and plutonium for safeguards and fissile material control for more than 20 years. The REIMEP programme (Regular European Inter-laboratory Measurement Evaluation Programme) serves as a tool for laboratories to demonstrate their abilities to measure uranium and plutonium isotopic ratios in a variety of sample forms, chosen where possible to be typical of fissile material samples commonly found in the nuclear industry and controlled by nuclear safeguards authorities. Previous REIMEP campaigns have included samples such as uranium oxide, uranium in nitric acid, uranium in the form of UF_6 [6,7], plutonium oxide, and others.

In October 2005 the REIMEP 18 campaign for the measurement of isotopic ratios of uranium in a simple nitric acid matrix was started. This campaign follows earlier campaigns for the measurement of uranium isotopic ratios, which turned out to be very valuable in showing the measurement capabilities of laboratories active in this measurement field. This field is not limited to the nuclear industry and laboratories working for nuclear safeguards. For REIMEP 18 a significant number of laboratories from the environmental, geochemistry and cosmo-chemistry areas were also included. The reason for this expansion was first to obtain a more representative general picture of measurement capabilities for uranium isotopic abundances and second to possibly expand the collaboration with scientists from these areas. The field of uranium isotopic measurements includes a variety of different measurement techniques. For measuring isotopic ratios various types of inductively coupled-plasma mass-spectrometry (ICP-MS) are widely used. Thermal-ionization mass-spectrometry (TIMS) is a well known technique and further methods such as accelerator mass-spectrometry (AMS), alpha spectrometry and selective laser ionization mass spectrometry are being used as well, focussing on specific isotope ratios. (A complete list of measurement techniques used is given in Table 1.) Between the various measurement techniques, various user groups and applications the requirements regarding precision and accuracy may differ significantly. However, in this paper all results are shown and discussed together without attaching any qualitative evaluation to any of the techniques.

For the REIMEP 18 campaign the uranium samples were provided in nitric acid solution, a matrix that is easy to handle and does not require extensive sample preparation steps, such as separation from other elements or purification. For this reason the REIMEP 18 campaign focussed on the ‘pure’ instrumental part of uranium isotope measurements by the technique the laboratory applied. But this measurement task already represents quite a challenge because of the large range of isotopic compositions for uranium. The amount of uranium provided for each sample was 2.5 mg, to allow an isotopic measurement without any constraint on the analytic

Table 1
Abbreviations of measurement techniques for Figs. 1–5

Abbreviation	Technique
AMS	Accelerator mass spectrometry
Alpha Spectrometry	Alpha spectrometry
HR-ICP-MS	High resolution inductively coupled-plasma mass spectrometry
ICP-IDMS	Isotope dilution inductively coupled-plasma mass spectrometry
ICP-QMS	Quadrupole inductively coupled-plasma mass spectrometry
MC-ICP-MS	Multi-collector inductively coupled-plasma mass spectrometry
SF-ICP-MS	Sector field inductively coupled-plasma mass spectrometry
LASER	Isotope selective laser ionization mass spectrometry
TIMS	Thermal ionisation mass spectrometry
TIMS TE	Thermal ionisation mass spectrometry using total evaporation

performance for all relevant techniques. The solutions only had to be diluted depending on the instrumental requirements.

Obviously these campaign samples are very much different from any environmental or forensic nuclear samples, both regarding the matrix and the amount of uranium. For uranium or plutonium measurements on environmental samples, which usually require considerable chemical preparation prior to mass spectrometry, IRMM also carries out the Nuclear Signatures Inter-laboratory Measurement Evaluation Programme (NUSIMEP). Several NUSIMEP rounds with different sample matrices have been organized within the last few years [8,9].

Invitations to participate were sent to a large number of laboratories. The response was very positive; registrations were received from 85 laboratories in 26 countries. Within the U.S., the New Brunswick Laboratory (NBL, U.S. DOE) acted as co-organizer by contacting a number of laboratories that regularly participate in NBL's measurement evaluation programme. All participating laboratories are active in the fields of research and development, measurement of radioactivity in the environment, monitoring of nuclear facilities, medical applications, measurements for fissile material control or safeguards. The entire participating community can be divided into two sections: in the first those doing research and development in scientific disciplines such as geochemistry (ca. 20 participants) and in the other those involved in nuclear isotope ratio measurements in the environment, in nuclear facilities or for fissile material control and safeguards purposes.

2. Sample preparation and certification measurements

The mass-spectrometric certification measurements for uranium isotope ratio measurements for the REIMEP 18 campaign were performed applying the latest knowledge of measurement methodology. This led to state-of-the-art precision and accuracy in isotopic measurements. Four samples of depleted to low-enriched uranium were selected from the IRMM stock. The original uranium samples were in UF_6 form. They were certified for the major ratio $n(^{235}U)/n(^{238}U)$ using a Varian MAT511 UF_6-gas source mass spectrometer (GSMS), calibrated using certified materials traceable to synthetic isotope mixtures. The samples in UF_6 form were hydrolyzed and calcined in order to obtain the uranium in oxide form (U_3O_8). The oxides were dissolved in nitric acid to obtain the batch solutions for REIMEP 18 A–D. In order to verify the certified major ratios $n(^{235}U)/n(^{238}U)$, thermal-ionization mass-spectrometer (TIMS) measurements were performed using the "Modified Total Evaporation" (MTE) technique as described in Ref. [10].

The minor uranium isotope ratios, $n(^{234}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$ were then measured and certified using a Triton TIMS. The method is described in detail in Refs. [10,11]. All $n(^{234}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$ ratios higher than ca. 5×10^{-5} were measured using Faraday collectors only, with current amplifiers that were equipped with $10^{12}\ \Omega$ resistors to improve the signal-to-noise ratio for the detection of ^{234}U and ^{236}U. All $n(^{236}U)/n(^{238}U)$ ratios below 5×10^{-5}, which only applied to samples REIMEP 18 A and D, were measured using an secondary electron multiplier (SEM) in combination with an energy filter for improved abundance sensitivity. This was inter-calibrated against the Faraday cups using the ^{234}U beam.

The batch solutions for REIMEP 18 A–D were sealed into ampoules containing 2.5 mg of uranium in 0.5 ml of 0.5 M nitric acid solution. The sample amounts were chosen in order to achieve a total alpha activity of less than 1000 Bq for each set of four samples, which allowed the sample sets to be shipped as non-nuclear material. This was a significant advantage for a large number of participating laboratories, because extensive administrative work related to nuclear transport requirements could be avoided. Finally additional verification measurements were performed using TIMS for all isotope ratios on one REIMEP 18 A–D sample set and showed good agreement with the certified values.

3. Results and discussion

Results for the $n(^{234}U)/n(^{238}U)$, $n(^{235}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$ ratios for samples REIMEP 18 A–D are presented in Figs. 1–5. In each graph the various measurement techniques used are indicated. The acronyms for the techniques are explained in Table 1. Each participant was allowed to check only one of the techniques in Table 1; meaning they are all exclusive, with the exception of the acronym "TIMS TE" which

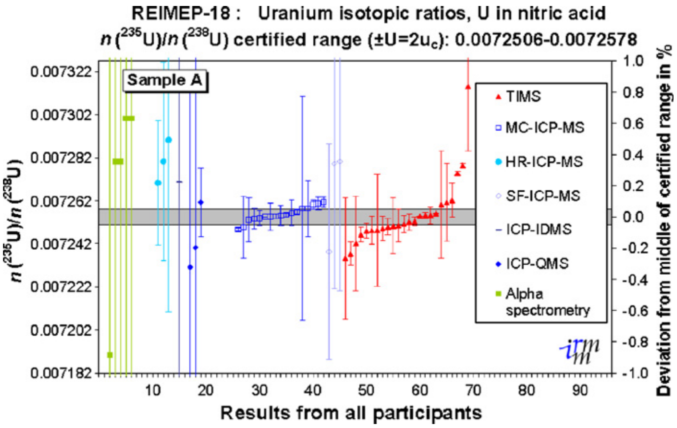

Fig. 1. Results for the $n(^{235}U)/n(^{238}U)$ ratio for REIMEP 18 A.

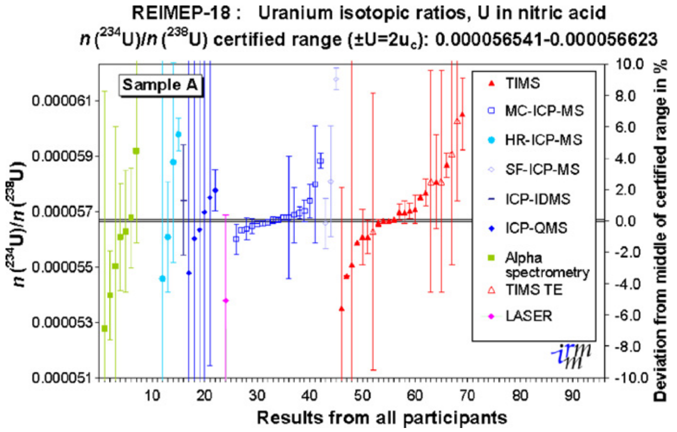

Fig. 2. Results for the $n(^{234}U)/n(^{238}U)$ ratio for REIMEP 18 A, using ±10% scale.

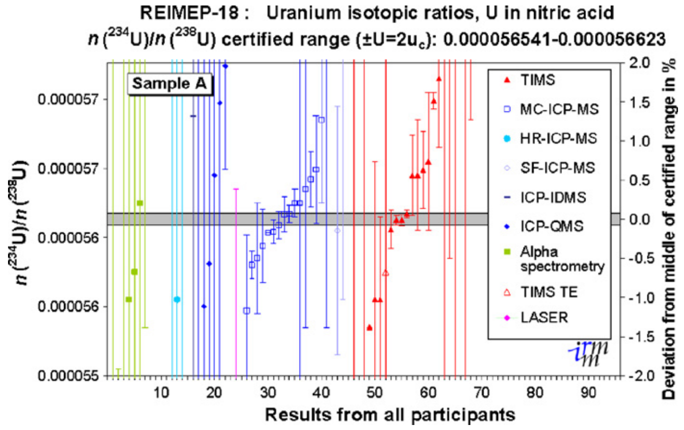

Fig. 3. Results for the $n(^{234}\mathrm{U})/n(^{238}\mathrm{U})$ ratio for REIMEP 18 A, using ±2% scale.

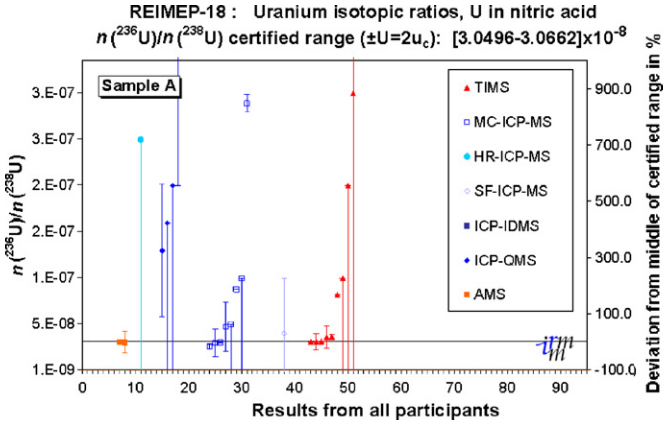

Fig. 4. Results for the $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ ratio for REIMEP 18 A.

means "TIMS Total Evaporation" and belongs to TIMS. Total evaporation is a special TIMS technique in which the sample is completely evaporated from the filament in order to minimize or sometimes even remove any mass fractionation effects.

A general observation for all ratios and all the samples is that the spread among the data increases with decreasing ratio values; this is simply related to the ion beam intensities, counting statistics and/or amplifier noise. For all techniques the spread of the data is significantly (at least 2–3 times) higher than the certified range, which is displayed in grey as $\pm k u_C$ with coverage factor $k = 2$. The relative certified range is ±0.05% for $n(^{235}\mathrm{U})/n(^{238}\mathrm{U})$, ±0.06–0.08% for $n(^{234}\mathrm{U})/n(^{238}\mathrm{U})$ and for $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ the range is ±0.05–0.27%, strongly depending on the value of the ratio.

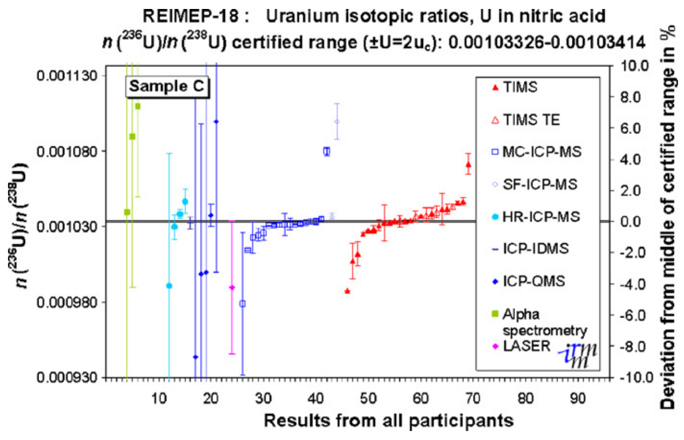

Fig. 5. Results for the $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ ratio for REIMEP 18 D.

The certified $n(^{235}\mathrm{U})/n(^{238}\mathrm{U})$ ratios of samples REIMEP 18 A–D lie between ca. 0.004 (depleted uranium) and 0.035 (low enriched uranium, e.g., see Fig. 1) and cover a range typical for nuclear safeguards samples. Sample REIMEP 18 A is quite close to natural uranium. From the $n(^{235}\mathrm{U})/n(^{238}\mathrm{U})$ results from all participants the following observations can be made:

1. The data spread depends on the applied techniques: multi-collector inductively coupled-plasma mass spectrometry (MC-ICP-MS) and TIMS show the smallest spread. For alpha spectrometry the results are between ±1% of the certified value only for sample REIMEP 18 A which has an isotopic composition close to natural. For all other samples the deviations are more than 1% and the data are therefore not shown on the graphs.
2. The uncertainties reported for many of the MC-ICP-MS and TIMS results seem to be quite small and appear to be underestimated for about 20% of the data. One possible reason might be the fact that several participants used natural uranium samples as a standard for the mass fractionation correction by using the consensus value of $n(^{238}\mathrm{U})/n(^{235}\mathrm{U}) = 137.88$ as "reference value". Although this number is well known and established in the literature, it is not a certified value and moreover, it does not have any (certified) uncertainty associated with it. As a consequence, the uncertainty contribution of this standard sample used for mass fractionation correction, which is in many cases the dominant contribution, is missing within the uncertainty budget calculation for the corrected ratio of the sample. This neglect can lead to a dramatic underestimation of the uncertainties; in several cases this neglect might have even caused an apparent deviation of a measured ratio from the certified value.

The certified $n(^{234}\mathrm{U})/n(^{238}\mathrm{U})$ ratios of samples REIMEP 18 A–D lie between ca. 0.000055 (close to natural uranium, see Fig. 2) and 0.00035 (slightly enriched uranium) and cover a range typical for nuclear safeguards samples. From the $n(^{234}\mathrm{U})/n(^{238}\mathrm{U})$ results the following observations can be made:

1. The data spread depends on the applied technique. Measurements done using MC-ICP-MS show the smallest spread, followed by TIMS, then other ICP-techniques, isotope selective laser ionization mass spectrometry and alpha spectrometry.
2. For $n(^{234}\mathrm{U})/n(^{238}\mathrm{U})$ ratios smaller than about 0.0001, which applies to the samples REIMEP 18 A and REIMEP 18 D, the TIMS-TE results (TE = total evaporation) have a tendency of being slightly higher than the expected value. This might be due to a neglect of the peak tailing correction to be done for the tailing of the major ion beams of $^{235}\mathrm{U}$ and $^{238}\mathrm{U}$.

3. Some of the reported uncertainties for results from MC-ICP-MS and TIMS seem to be quite small and may be underestimated. This is clearly visible in Fig. 3, where the results for REIMEP 18 A are shown with a vertical scale of ±2%. Possible reasons might be:
    - (a) Use of a natural consensus standard for the mass fractionation correction using the $n(^{235}\text{U})/n(^{238}\text{U})$ ratio. In this case the uncertainty of the corrected $n(^{235}\text{U})/n(^{238}\text{U})$ ratio is underestimated, leading to an incomplete uncertainty budget calculation for $n(^{234}\text{U})/n(^{238}\text{U})$.
    - (b) Underestimation or neglect of uncertainty contributions arising from detector inter-calibration (e.g., secondary electron multiplier versus Faraday cups) or linearity correction of detectors, especially secondary electron multipliers.

The certified values of $n(^{236}\text{U})/n(^{238}\text{U})$ for the REIMEP 18 samples lie between ca. 10^{-8} (closest to natural uranium, see Fig. 4) and 0.001 (similar to processed uranium, see Fig. 5). From the $n(^{236}\text{U})/n(^{238}\text{U})$ results the following observations can be made:

1. The data spread depends on the techniques applied and on the order of magnitude of the ratio. For samples REIMEP 18 B and C with $n(^{236}\text{U})/n(^{238}\text{U}) > 0.0001$, measurements performed using MC-ICP-MS show the smallest spread, followed by TIMS, other ICP-techniques and alpha-spectrometry. For samples REIMEP 18 D and A, with ratios of the order of 10^{-7} and 3×10^{-8}, AMS (only two results) and TIMS show the smallest spread, followed by the ICP-MS techniques.
2. For samples REIMEP 18 B and C with ratios of 0.0003 and 0.001, respectively, most of the TIMS-TE results (TE = total evaporation) have a tendency of being slightly higher than the expected value. These deviations are very probably due to neglecting the peak tailing correction due to the large ion beam at mass 238. More care has to be taken for accurate measurements of $n(^{236}\text{U})/n(^{238}\text{U})$ when measured in simple static total evaporation mode.
3. The results for sample A, with $n(^{236}\text{U})/n(^{238}\text{U}) = 3 \times 10^{-8}$ is shown using a range from −100% to +1000% in Fig. 4. Clearly isotopic measurements within this extreme dynamic range of 7–8 orders of magnitude still constitute a great challenge for the instrument and operator. The reported uncertainties seem often to be underestimated. Possibly not all uncertainty components, such as tailing effects, instrumental background, detector inter-calibration, etc., are considered sufficiently. The ICP techniques are at a disadvantage because of the larger tailing contributions compared with TIMS and AMS, even when an energy filter is used.
4. In order to provide a proof for the reliability of the certified values of $n(^{236}\text{U})/n(^{238}\text{U})$ around 10^{-7} to 10^{-8} for samples REIMEP 18 D and A, a separate verification measurement series of the new IRMM-075 synthetic isotope mixtures with $n(^{236}\text{U})/n(^{238}\text{U})$ ratios of 10^{-4}, 10^{-5}, 10^{-6}, 10^{-7}, 10^{-8} and 10^{-9} was performed [12,13]. The $n(^{236}\text{U})/n(^{238}\text{U})$ ratios of these standard samples were measured using the IRMM-TIMS procedure and showed excellent agreement with the certified ratios, as shown in Fig. 6.

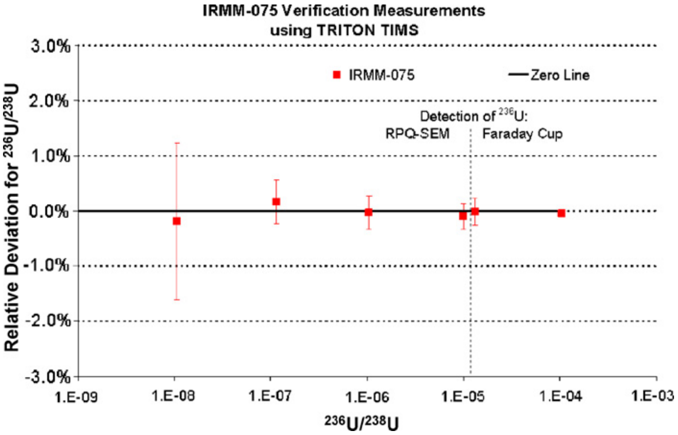

Fig. 6. Comparison between the $n(^{236}\text{U})/n(^{238}\text{U})$ ratios measured using the Triton TIMS and the certified values for IRMM-075. Each data point represents the average of five measurements.

4. Evaluation of the questionnaire

The submission of the measured data was accompanied by a questionnaire to be completed by each participating laboratory. The most important results are presented briefly here, mainly focussed on mass spectrometry issues.

About 85% of the laboratories using mass spectrometry applied a correction for mass fractionation using a reference material. Only one laboratory applied an internal correction using a $^{233}\text{U}/^{236}\text{U}$ double spike, the majority performed an external correction using a standard or reference material. In some cases even two bracketing standards were used.

Because uranium isotope abundances cover a large dynamic range, many of the participating laboratories used a combination of different detectors for the uranium isotopes ^{234}U, ^{235}U, ^{236}U and ^{238}U. Most common are Faraday cups and various types of secondary electron multipliers (SEMs), either discrete dynode or continuous dynode multipliers. SEMs are the preferred detectors for the minor isotopes ^{234}U and ^{236}U and Faraday cups are predominantly used for the major isotopes ^{235}U and ^{238}U.

The inter-calibration between different detectors such as Faraday cups and various types of SEMs is an important part of the measurement procedure. Most of the laboratories (55%) use standards to achieve the inter-calibration, either using one standard or even two standards bracketing the unknown sample ratio. But a lot of laboratories (45%) prefer to use an ion beam of the (same) sample to cross-calibrate the SEM against the Faraday cups. Some of the laboratories even apply an internal calibration between SEM and Faraday cup regularly throughout the sample measurement. The cross-calibration approach can have the advantage of taking into account any run-to-run variations of the calibration factor (run of either a sample or a standard, “external” procedure), or even within-run variations (“internal” procedure). The inter-calibration between an SEM against Faraday cups plays an important role for the measurement of the minor uranium ratios $n(^{234}\text{U})/n(^{238}\text{U})$ and $n(^{236}\text{U})/n(^{238}\text{U})$, for which only few reliable certified standards with low uncertainties exist [10,14].

Each detector, Faraday cup or secondary electron multiplier (SEM), has to be checked for linearity. Usually Faraday cups do not show any deviation from linearity, which can be easily confirmed by measurements of suitable isotopic reference materials such as IRMM-072, IRMM-073 or IRMM-074 [15,16]. In contrast, SEM detection systems usually exhibit non-linearity effects. Each SEM system operated in pulse counting mode consists of the SEM detector itself and a pulse amplifier and counter. The pulse amplifier always has a certain dead time, which is the time duration after each pulse for which the amplifier cannot accept any further count. In the first approximation the dead time correction is linear with the count rate and has been described in the literature many times. But recently, additional non-linearity effects have also been observed and investigated which originate from the SEM detector itself and require an additional correction [17,18]. It seems only a few laboratories make a distinction between the dead time effect and additional effects of the SEM detector and in many cases these effects are either not taken into account or not fully investigated. But the results from REIMEP 18, especially for the minor uranium ratios $n(^{234}\mathrm{U})/n(^{238}\mathrm{U})$ and $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ demonstrate that non-linearity effects or the lack of proper corrections can still cause significant inaccuracies in the measurements of these ratios.

5. Conclusions

The REIMEP 18 inter-laboratory comparison campaign for uranium isotope measurements was a success. The overall response of 85 registered and 71 actually participating laboratories was exceptionally high. The results of this campaign may therefore be considered as a representative picture of present uranium isotopic measurement capabilities for a broad range of disciplines and on a worldwide scale. The REIMEP 18 campaign samples are being applied as quality control samples by several laboratories and even used to demonstrate the analytical performance in publications [19,20]. This campaign was focused on the ‘pure’ measurement of uranium isotopic abundances rather than on the full analytical sample analysis procedure including, e.g., chemical sample preparation. The outcome of this campaign is therefore specific to the instrumentation utilized to measure uranium isotopic ratios, to the various measurement procedures, the calibration applied and correction strategies. From the results the following main conclusions can be drawn:

1. For measurements of uranium isotopic ratios a variety of different measurement techniques is in use. Most prominent is mass spectrometry, and among different types of mass spectrometry TIMS and various types of ICP-MS, especially MC-ICP-MS, are the most frequently used.
2. Each technique has its limitations, e.g., alpha-spectrometry and isotope selective laser ionization mass spectrometry are preferentially used for certain isotope ratios or specific ranges of ratios. ICP-MS has limited capabilities for measurements of ratios covering a large dynamic range, e.g., for $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ ratios of the order of 10^{-7} and below. For this type of measurement TIMS and AMS show the best performance.
3. For TIMS measurements, the performance for routine nuclear safeguards measurements of the minor isotope ratios, e.g., using the total evaporation technique, could be improved. It seems that the capabilities provided by modern TIMS instruments are not always fully applied in order to reach the best possible performance of TIMS. Plans for improvement have been proposed by IRMM, e.g., by an expanded implementation of the “modified total evaporation” technique into the standard software of modern TIMS instruments [10].
4. Corrections for effects such as mass fractionation or detector non-linearity are usually performed using known isotopic standards. Many laboratories use certified isotope reference materials provided by e.g., NIST/NBL or IRMM, but quite a large number also uses “consensus” type standards of natural uranium. The advantage of the better availability is often compromised by the lack of complete uncertainty propagation, leading to underestimated uncertainties and possibly biased results. There is an obvious need for more discussion and inter-laboratory knowledge exchange about guidelines for calculating uncertainties for isotope ratio measurements.

It is planned to organize measurement campaigns such as REIMEP 18 on a regular basis in order to re-assess the status of uranium isotope measurement capabilities, also to fulfil requirements for external quality control and to address upcoming measurement problems.

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