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JRC REFERENCE MATERIALS REPORT

Preparation and Certification of Highly Enriched Uranium Nitrate Solutions IRMM-3000 Series

Certified for Isotope Ratios

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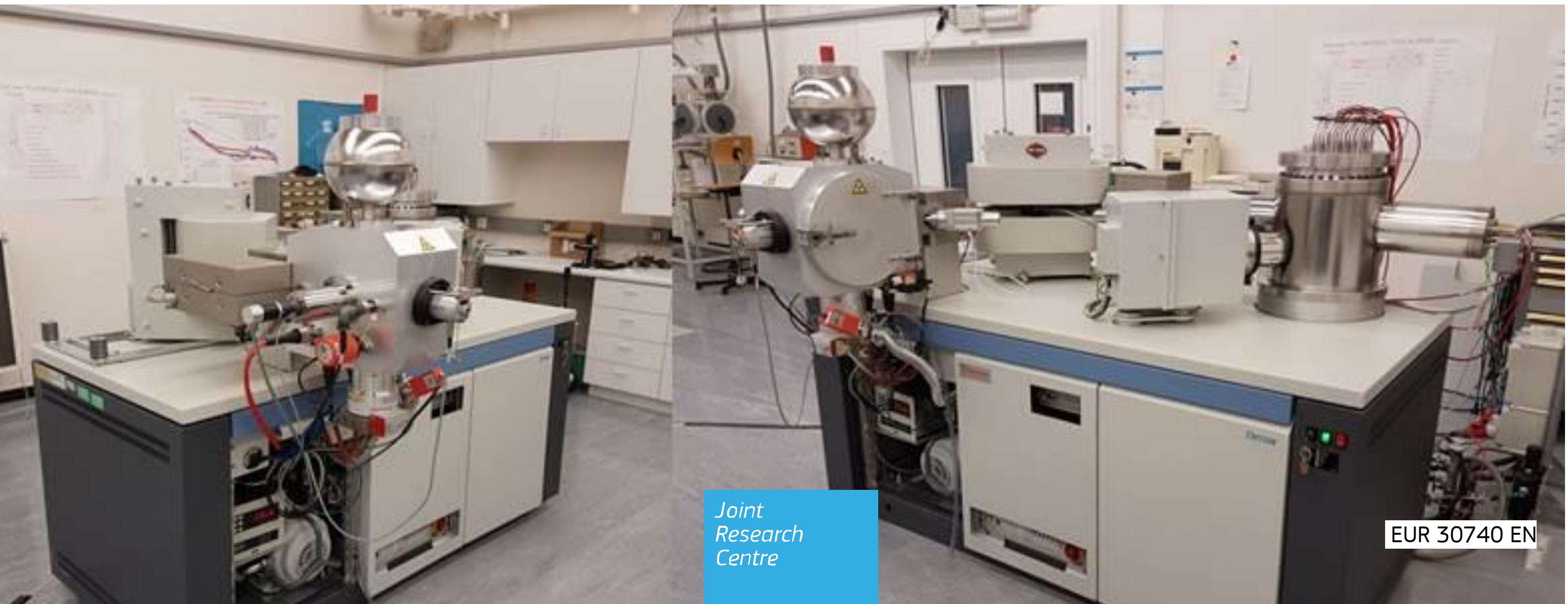

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Contents

Foreword

The Directorate G "Nuclear Safety and Security", Unit G.2 "Standards for Nuclear Safety, Security and Safeguards" (SN3S) at the European Commission's Joint Research Centre (JRC) in Geel, Belgium (formerly known as the "Institute for Reference Materials and Measurements" IRMM), provides a wide range of nuclear Certified Reference Materials (CRMs) to safeguards authorities and the nuclear industry.

This report describes the certification of IRMM-3000, a series of highly enriched uranium nitrate solution isotopic reference materials. The project was a collaboration of JRC-G.2 with the JRC-G.II.8 (Analytical Service) in Karlsruhe/Germany, the International Atomic Energy Agency (IAEA), in Seibersdorf Austria, and the Oak Ridge National Laboratory (ORNL), in Oak Ridge, USA.

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Abstract

This report describes the certification of the IRMM-3000 series of uranium nitrate solution reference materials, certified for the uranium isotopic composition. The certified values and their uncertainties were assigned following ISO 17034 [1], ISO Guide 35 [2] and the Guide to the Expression of Uncertainty in Measurement [3].

This certification project was a collaboration between the International Atomic Energy Agency (IAEA-SGAS, Austria), the Oak Ridge National Laboratory (ORNL, U.S. DOE), Lawrence Livermore National Laboratory (LLNL, U.S.DOE) and the Joint Research Centre (JRC-Geel, Unit G.2 in Geel/Belgium and Unit G.II.8 in Karlsruhe/Germany).

The IRMM-3000-series consists of a set of five highly enriched uranium nitrate solution reference materials named IRMM-3020, IRMM-3035, IRMM-3050, IRMM-3075 and IRMM-3090 with the isotopic ratio $n(^{235}U)/n(^{238}U)$ varying from about 0.25 to 6.6, and the ^{235}U enrichment varying from 20 % to 90 %. The solutions were prepared by dissolving uranium oxide (U_3O_8) materials, which were prepared by gravimetrically mixing oxides which were highly enriched in ^{235}U and ^{238}U, in a similar way as performed for the preparation of the IRMM-074, IRMM-075 and IRMM-3636a,b series of isotope reference materials [4,5,6].

The isotopically enriched starting materials were purified using identical methods involving separation on anion and cation columns followed by a precipitation as peroxide. Both starting materials were treated in separate clean glove-boxes to eliminate cross-contamination. The oxides were calcinated in parallel to convert them into U_3O_8, in an oven installed in a glove-box that provided a controlled low-humidity environment. For each of the mixtures, the oxides of ^{235}U and ^{238}U were weighed and dissolved to form a solution with a mass fraction of about 5 mg /g. The final solutions were dispensed into individual quartz ampoules containing 0.5 mL, 1 mL or 5 mL of a 1 mol/L nitric acid solution and subsequently flame-sealed.

The $n(^{235}U)/n(^{238}U)$ isotope ratios of the IRMM-3000-series were calculated based on the weighings of the two starting materials enriched in ^{235}U and ^{238}U and the measured impurities of both. The main contributions to the final uncertainties of the isotopic ratios are originating from the weighing uncertainties, the uncertainties of the measured isotope ratios and impurities in the two starting materials. The $n(^{234}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$ isotope ratios of the IRMM-3000-series were certified by TIMS/MTE at JRC-Geel.

In this report, the methods for the preparation via gravimetry and certification of the IRMM-3000 series will be described, including verification measurements for the isotope ratios by several methods of mass spectrometry, such as TIMS/MTE, TIMS/DS and MC-ICPMS. As explained in detail in the report, in the case of IRMM-3035, the isotope ratios were only certified using mass spectrometry by TIMS/MTE and TIMS/DS at JRC-Geel, with subsequent verification using TIMS/MTE and MC-ICPMS by the IAEA and ORNL.

The isotopic reference materials IRMM-3020 (20% ^{235}U), IRMM-3035 (35% ^{235}U), IRMM-3050 (50% ^{235}U), IRMM-3075 (75% ^{235}U) and IRMM-3090 (90% ^{235}U) are part of a systematic program of JRC-G.2 to supply isotope reference materials to the safeguards community. The materials are intended for the calibration of instruments and methods, quality control purposes. As with any certified reference material, they can also be used for validation studies.

The following values were assigned to the IRMM-3020:

URANIUM NITRATE SOLUTION IRMM-3020		
	Isotope amount ratio	
	Certified value [1)] [mol/mol]	Uncertainty [2)] [mol/mol]
$n(^{234}U)/n(^{238}U)$	0.000004931	0.000000035
$n(^{235}U)/n(^{238}U)$	0.254264	0.000041
$n(^{236}U)/n(^{238}U)$	0.000009805	0.000000063
	Isotope amount fraction	
	Certified value [3)] [mol/mol]	Uncertainty [2)] [mol/mol]
$n(^{234}U)/n(U)$	0.000003931	0.000000028
$n(^{235}U)/n(U)$	0.202717	0.000026
$n(^{236}U)/n(U)$	0.000007818	0.000000050
$n(^{238}U)/n(U)$	0.797271	0.000026
	Isotope mass fraction	
	Certified value [3) 4)] [g/g]	Uncertainty [2)] [g/g]
$m(^{234}U)/m(U)$	0.000003875	0.000000027
$m(^{235}U)/m(U)$	0.200670	0.000026
$m(^{236}U)/m(U)$	0.000007772	0.000000050
$m(^{238}U)/m(U)$	0.799318	0.000026
	Molar mass	
	Certified value [3) 4)] [g/mol]	Uncertainty [2)] [g/mol]
$M(U)$	237.441214	0.000078

1) The certified values are traceable to the International System of units (SI) via gravimetrical preparation.

2) The uncertainty is the expanded uncertainty with a coverage factor $k = 2$ corresponding to a level of confidence of about 95 % estimated in accordance with ISO/IEC Guide 98-3, Guide to the Expression of Uncertainty in Measurement (GUM:1995), ISO, 2008.

3) These values are calculated using the isotope amount ratios and therefore traceable to the SI. The calculation of n(U), m(U) and M(U) includes the contributions from the isotopes ^{234}U, ^{235}U, ^{236}U and ^{238}U.

4) These values are calculated using the values listed below from *Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003:*

$M(^{234}U)$ = 234.0409504 ± 0.0000024 g·mol^{-1} (k = 2)

$M(^{235}U)$ = 235.0439282 ± 0.0000024 g·mol^{-1} (k = 2)

$M(^{236}U)$ = 236.0455662 ± 0.0000024 g·mol^{-1} (k = 2)

$M(^{238}U)$ = 238.0507870 ± 0.0000032 g·mol^{-1} (k = 2)

The following values were assigned to the IRMM-3035:

URANIUM NITRATE SOLUTION IRMM-3035		
	Isotope amount ratio	
	Certified value [1)] [mol/mol]	Uncertainty [2)] [mol/mol]
$n(^{234}U)/n(^{238}U)$	0.000010669	0.000000069
$n(^{235}U)/n(^{238}U)$	0.548531	0.000089
$n(^{236}U)/n(^{238}U)$	0.000021224	0.000000086
	Isotope amount fraction	
	Certified value [3)] [mol/mol]	Uncertainty [2)] [mol/mol]
$n(^{234}U)/n(U)$	0.000006890	0.000000045
$n(^{235}U)/n(U)$	0.354220	0.000037
$n(^{236}U)/n(U)$	0.000013706	0.000000056
$n(^{238}U)/n(U)$	0.645760	0.000037
	Isotope mass fraction	
	Certified value [3) 4)] [g/g]	Uncertainty [2)] [g/g]
$m(^{234}U)/m(U)$	0.000006804	0.000000044
$m(^{235}U)/m(U)$	0.351317	0.000037
$m(^{236}U)/m(U)$	0.000013651	0.000000056
$m(^{238}U)/m(U)$	0.648662	0.000037
	Molar mass	
	Certified value [3) 4)] [g/mol]	Uncertainty [2)] [g/mol]
$M(U)$	236.98564	0.00011

1) The certified values are traceable to the International System of units (SI) via IRMM-3636a.

2) The uncertainty is the expanded uncertainty with a coverage factor $k = 2$ corresponding to a level of confidence of about 95 % estimated in accordance with ISO/IEC Guide 98-3, Guide to the Expression of Uncertainty in Measurement (GUM:1995), ISO, 2008.

3) These values are calculated using the isotope amount ratios and therefore traceable to the SI. The calculation of $n(U)$, $m(U)$ and M(U) includes the contributions from the isotopes ^{234}U, ^{235}U, ^{236}U and ^{238}U.

4) These values are calculated using the values listed below from *Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003:*

$M(^{234}U)$ = 234.0409504 ± 0.0000024 g·mol^{-1} (k = 2)

$M(^{235}U)$ = 235.0439282 ± 0.0000024 g·mol^{-1} (k = 2)

$M(^{236}U)$ = 236.0455662 ± 0.0000024 g·mol^{-1} (k = 2)

$M(^{238}U)$ = 238.0507870 ± 0.0000032 g·mol^{-1} (k = 2)

The following values were assigned to the IRMM-3050:

URANIUM NITRATE SOLUTION IRMM-3050		
	Isotope amount ratio	
	Certified value [1) [mol/mol]	Uncertainty [2) [mol/mol]
$n(^{234}U)/n(^{238}U)$	0.00001991	0.00000013
$n(^{235}U)/n(^{238}U)$	1.02359	0.00018
$n(^{236}U)/n(^{238}U)$	0.00003969	0.00000012
	Isotope amount fraction	
	Certified value [3) [mol/mol]	Uncertainty [2) [mol/mol]
$n(^{234}U)/n(U)$	0.000009838	0.000000063
$n(^{235}U)/n(U)$	0.505814	0.000044
$n(^{236}U)/n(U)$	0.000019615	0.000000060
$n(^{238}U)/n(U)$	0.494157	0.000044
	Isotope mass fraction	
	Certified value [3) 4)] [g/g]	Uncertainty [2) [g/g]
$m(^{234}U)/m(U)$	0.000009735	0.000000063
$m(^{235}U)/m(U)$	0.502636	0.000044
$m(^{236}U)/m(U)$	0.000019574	0.000000060
$m(^{238}U)/m(U)$	0.497335	0.000044
	Molar mass	
	Certified value [3) 4)] [g/mol]	Uncertainty [2) [g/mol]
M(U)	236.52980	0.00013

1) The certified values are traceable to the International System of units (SI) via gravimetrical preparation.

2) The uncertainty is the expanded uncertainty with a coverage factor $k = 2$ corresponding to a level of confidence of about 95 % estimated in accordance with ISO/IEC Guide 98-3, Guide to the Expression of Uncertainty in Measurement (GUM:1995), ISO, 2008.

3) These values are calculated using the isotope amount ratios and therefore traceable to the SI. The calculation of n(U), m(U) and M(U) includes the contributions from the isotopes ^{234}U, ^{235}U, ^{236}U and ^{238}U.

4) These values are calculated using the values listed below from *Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003:*

$M(^{234}U)$ = 234.0409504 ± 0.0000024 g·mol^{-1} (k = 2)

$M(^{235}U)$ = 235.0439282 ± 0.0000024 g·mol^{-1} (k = 2)

$M(^{236}U)$ = 236.0455662 ± 0.0000024 g·mol^{-1} (k = 2)

$M(^{238}U)$ = 238.0507870 ± 0.0000032 g·mol^{-1} (k = 2)

The following values were assigned to the IRMM-3075:

URANIUM NITRATE SOLUTION IRMM-3075		
	Isotope amount ratio	
	Certified value 1) [mol/mol]	Uncertainty 2) [mol/mol]
$n(^{234}\text{U})/n(^{238}\text{U})$	0.00005473	0.00000035
$n(^{235}\text{U})/n(^{238}\text{U})$	2.81872	0.00044
$n(^{236}\text{U})/n(^{238}\text{U})$	0.00010966	0.00000020
	Isotope amount fraction	
	Certified value 3) [mol/mol]	Uncertainty 2) [mol/mol]
$n(^{234}\text{U})/n(\text{U})$	0.000014332	0.000000091
$n(^{235}\text{U})/n(\text{U})$	0.738100	0.000030
$n(^{236}\text{U})/n(\text{U})$	0.000028716	0.000000053
$n(^{238}\text{U})/n(\text{U})$	0.261857	0.000030
	Isotope mass fraction	
	Certified value 3) 4) [g/g]	Uncertainty 2) [g/g]
$m(^{234}\text{U})/m(\text{U})$	0.000014224	0.000000091
$m(^{235}\text{U})/m(\text{U})$	0.735636	0.000030
$m(^{236}\text{U})/m(\text{U})$	0.000028742	0.000000053
$m(^{238}\text{U})/m(\text{U})$	0.264321	0.000030
	Molar mass	
	Certified value 3) 4) [g/mol]	Uncertainty 2) [g/mol]
$M(\text{U})$	235.831309	0.000091

1) The certified values are traceable to the International System of units (SI) via gravimetrical preparation.

2) The uncertainty is the expanded uncertainty with a coverage factor $k = 2$ corresponding to a level of confidence of about 95 % estimated in accordance with ISO/IEC Guide 98-3, Guide to the Expression of Uncertainty in Measurement (GUM:1995), ISO, 2008.

3) These values are calculated using the isotope amount ratios and therefore traceable to the SI. The calculation of n(U), m(U) and M(U) includes the contributions from the isotopes ^{234}U, ^{235}U, ^{236}U and ^{238}U.

4) These values are calculated using the values listed below from *Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003:*

$M(^{234}U)$ = 234.0409504 ± 0.0000024 g·mol^{-1} (k = 2)

$M(^{235}U)$ = 235.0439282 ± 0.0000024 g·mol^{-1} (k = 2)

$M(^{236}U)$ = 236.0455662 ± 0.0000024 g·mol^{-1} (k = 2)

$M(^{238}U)$ = 238.0507870 ± 0.0000032 g·mol^{-1} (k = 2)

The following values were assigned to the IRMM-3090:

URANIUM NITRATE SOLUTION IRMM-3090		
	Isotope amount ratio	
	Certified value 1) [mol/mol]	Uncertainty 2) [mol/mol]
$n(^{234}U)/n(^{238}U)$	0.00012770	0.00000083
$n(^{235}U)/n(^{238}U)$	6.57731	0.00092
$n(^{236}U)/n(^{238}U)$	0.00025654	0.00000034
	Isotope amount fraction	
	Certified value 3) [mol/mol]	Uncertainty 2) [mol/mol]
$n(^{234}U)/n(U)$	0.00001685	0.00000011
$n(^{235}U)/n(U)$	0.867983	0.000016
$n(^{236}U)/n(U)$	0.000033854	0.000000046
$n(^{238}U)/n(U)$	0.131966	0.000016
	Isotope mass fraction	
	Certified value 3) 4) [g/g]	Uncertainty 2) [g/g]
$m(^{234}U)/m(U)$	0.00001675	0.00000011
$m(^{235}U)/m(U)$	0.866520	0.000016
$m(^{236}U)/m(U)$	0.000033941	0.000000046
$m(^{238}U)/m(U)$	0.133429	0.000016
	Molar mass	
	Certified value 3) 4) [g/mol]	Uncertainty 2) [g/mol]
$M(U)$	235.440749	0.000048

1) The certified values are traceable to the International System of units (SI) via gravimetrical preparation.

2) The uncertainty is the expanded uncertainty with a coverage factor $k = 2$ corresponding to a level of confidence of about 95 % estimated in accordance with ISO/IEC Guide 98-3, Guide to the Expression of Uncertainty in Measurement (GUM:1995), ISO, 2008.

3) These values are calculated using the isotope amount ratios and therefore traceable to the SI. The calculation of $n(U)$, $m(U)$ and M(U) includes the contributions from the isotopes ^{234}U, ^{235}U, ^{236}U and ^{238}U.

4) These values are calculated using the values listed below from *Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003:*

$M(^{234}U)$ = 234.0409504 ± 0.0000024 g·mol^{-1} (k = 2)

$M(^{235}U)$ = 235.0439282 ± 0.0000024 g·mol^{-1} (k = 2)

$M(^{236}U)$ = 236.0455662 ± 0.0000024 g·mol^{-1} (k = 2)

$M(^{238}U)$ = 238.0507870 ± 0.0000032 g·mol^{-1} (k = 2)

1. Introduction

The Directorate G "Nuclear Safety and Security", Unit G.2 "Standards for Nuclear Safety, Security and Safeguards" (SN3S) at the European Commission's Joint Research Centre, Geel in Belgium (formerly known as the "Institute for Reference Materials and Measurements" (IRMM)), provides a wide range of nuclear Certified Reference Materials (CRMs) to the safeguards authorities and the nuclear industry. This is an obligation under the Euratom treaty, where the need for isotope standards is explicitly mentioned, acknowledging their importance for the measurements of nuclear materials. For accurate mass spectrometric measurements in nuclear material accountancy and nuclear safeguards, suitable CRMs are needed to validate measurement procedures and to calibrate instruments.

In particular, for mass spectrometry, CRMs are needed which have been prepared and characterized using procedures which are independent on mass spectrometric techniques as much as possible. One possible way to achieve this, is to use highly isotopically enriched starting materials and mix them under gravimetrical control in the correct proportions to achieve given targets for the isotope ratios. This procedure has been applied for the preparation of uranium and plutonium isotope CRMs for many years by several institutions, like NIST, NBL and IRMM/JRC-Geel. In the 1980s, the IRMM-072 series [7] of isotope mixtures containing ^{233}U, ^{235}U and ^{238}U in various proportions was prepared based on this approach/method. About 20 years later, the isotope mixing program was restarted, and several further isotope CRMs were prepared and certified: the IRMM-074 [4] and IRMM-075 series [5], and the IRMM-3636a double spike [6].

In 2009, the IRMM-3000 series was prepared. It consists of a set of five highly enriched uranium nitrate solution reference materials named IRMM-3020, IRMM-3035, IRMM-3050, IRMM-3075 and IRMM-3090 with ^{235}U enrichments varying from 20 % to 90 %. The solutions were prepared by dissolving uranium oxide (U_3O_8) materials, which were prepared by gravimetrically mixing oxides that are highly enriched in ^{235}U and ^{238}U. However, the verification measurements by TIMS showed some significant differences from the calculations based on the gravimetrical preparation. It was suspected that incomplete impurity analyses of the ^{238}U starting material could be the cause for most of the differences, but this could not be confirmed at that time. Recently, the certification project for the IRMM-3000 series was restarted, including new mass spectrometric analyses of the isotope ratios performed at several laboratories and new impurity analysis for the ^{238}U starting material.

2. Preparation and Characterization of the IRMM-3000 series

Following the methods successfully applied in previous preparations [4,5,6], the IRMM-3000 series mixtures were made gravimetrically in 2009 by weighing purified, highly enriched oxides and mixing them in the correct proportions and subsequently dissolving the mixed oxides in nitric acid. The critical points of the preparation are as follows:

- Both enriched isotopes were handled completely isolated from each other to avoid cross-contamination (to maintain isotopic integrity).
- Exactly the same chemical purification procedure was used (and the same reagents) for both enriched starting materials in parallel.
- The purified starting materials were dried and then calcinated under the same conditions in terms of temperature and humidity within an oven built specifically for this purpose.
- A minimum weight of oxide of 200 mg was applied to keep the uncertainties from the weighing procedure as low as possible.

2.1 Purification of the enriched ^{235}U and ^{238}U starting materials

Both starting materials were submitted to a purification procedure as used in earlier prepared synthetic mixtures of uranium [4,5,6]. At that earlier time, experiments were made on the purification of the enriched isotopic uranium materials, using natural uranium for the tests. These investigations focused on the purification of uranium by anion and cation exchange and precipitation. Each of the steps of the purification cycles was evaluated by measuring a wide range of impurity elements semi-quantitatively by ICP-MS. The results of these experiments showed that three steps gave a satisfactory level of purification: anion exchange in nitric acid medium, cation exchange in HNO_3/THF (=tetrahydrofuran) and precipitation as peroxide.

The enriched starting materials were purified using identical methods and chemicals, but in separate clean glove-boxes to avoid cross-contamination. After completion of one step of the separation and before starting the next sequence of the procedure, the glove box was cleaned thoroughly and all unnecessary chemicals and glassware were removed.

2.1.1 Anion exchange

The uranium oxide with a mass of about 3 g – 4 g was dissolved in concentrated nitric acid, evaporated to dryness, subsequently dissolved in 7.5 mol/L nitric acid and evaporated to dryness twice. This was to ensure complete transformation into uranyl nitrate. The anion exchange resin used for this separation was BIORAD AG1X4, 100-200 mesh. The resin was washed to remove fines and slurried into a quartz column of diameter 0.8 cm and height 15 cm with a sintered quartz filter and a PTFE tap. The resin was conditioned by washing with 7.5 M nitric acid until it was chlorine free. The uranyl nitrate was dissolved in 10 mL of 7.5 mol/L nitric acid and this solution was pipetted onto the resin. The flask was rinsed three times with 2.5 mL 7.5 mol/L nitric acid and the rinse solutions were added to the column. The resin was washed with 40 mL 7.5 mol/L nitric acid; the uranium fraction was eluted with 385 mL of 7.5 mol/L nitric acid and evaporated till dryness for further purification over a cation exchange.

2.1.2 Cation exchange

As for the previous step, a quartz column of diameter 0.8 cm and height 15 cm with a sintered quartz filter at the bottom to retain the resin and a PTFE tap was used. The cation exchange resin was Bio-Rad AG 50Wx8, 100 to 200 mesh. The uranyl nitrate was dissolved in 10 mL of a mixture of 90% THF -10% 6 mol/L HNO_3 and the column was conditioned with the same solution. The solution was pipetted onto the resin, the flask was rinsed 3 times with 5 mL of 90% THF -10% 6 mol/L HNO_3 and the rinses added to the column. The uranium was washed through the column with 100 ml of 90% THF -10% 6 mol/L HNO_3 in order to retain the impurities. The solution was evaporated to dryness, dissolved in 10 mL concentrated nitric acid and again evaporated to dryness. Finally the residue was dissolved in 10 mL of 7.5 mol/L nitric acid.

2.1.3 Peroxide precipitation

The solution was adjusted to pH = 2 by drop-wise adding a 1:1 diluted solution of ammonium hydroxide, cooled down to 8 °C and kept for 24 hours whilst stirring. 75 mL of 30% hydrogen peroxide were added and the pH was kept constant at 2.0 by adding ammonium hydroxide (pH < 1.5) or nitric acid (pH > 2.5) where necessary. The solution was stirred further for another hour. A yellow-white precipitate was formed and allowed to settle. The supernatant was decanted off, the precipitate was washed with sub-boiled water containing hydrogen peroxide adjusted to pH 2. The precipitate was allowed to settle overnight at a temperature of 8 °C and subsequently filtered through a 47 mm diameter Millipore polycarbonate filter, pore size 0.4 µm in a borosilicate holder applying a slight under pressure. The precipitate was again rinsed 5 times with 2.5 mL sub-boiled water (containing hydrogen peroxide and adjusted to pH 2). The ammonium diuranate $(NH_4)_2U_2O_7$ precipitate was allowed to dry in air.

2.1.4 Equipment and chemicals

All metallic pieces on equipment used in the glove boxes were covered with a plastic coating. A ceramic hotplate and a quartz heater were used to evaporate solutions and dry the uranyl nitrate. All special labware used was made of quartz, FEP or PFA and were cleaned according to the standard cleaning procedure of the Ultra Clean Chemical Laboratory. Dust-free wiping tissues were used. All columns, Teflon bottles and quartz beakers were cleaned with nitric acid. Compared to the previous isotopic mixtures, higher specification reagents were used and highly pure (sub-boiled or commercially equivalent) water was routinely available. Moreover, the availability of ICP-MS at that time for measurement of trace amounts of impurities allowed for the purity of reagents and lab-ware to be controlled very efficiently.

2.2 Calcination

At the end of the purification process, for the ^{238}U starting material (BC02677) about 3.7 g of uranium and for the ^{235}U starting material (BC02154) about 3.3 g of uranium were available for calcination. In order to achieve the same stoichiometry, the oxides were calcinated simultaneously in an oven which was installed in one section of a stainless-steel glove-box with humidity control specially designed and constructed for this purpose The individual oxides were calcinated together in quartz crucibles held in a quartz housing to allow movement of air above the oxides but separating each material from the next. A programmed heating cycle was applied to calcine the oxides simultaneously. The temperature was slowly raised to 920 °C [4] and kept there for at least two hours before allowing to cool slowly. The purpose of this procedure was to form U_3O_8 and to ensure that each of the enriched materials have the same stoichiometry as much as possible. The glove-box environment was completely controlled and the humidity in the glove-box and oven were kept at less than 30 mg water per kg air throughout the final calcination process.

2.3 Mass metrology

The humidity was also controlled in the second section of the glove-box, in which two analytical balances were installed for weighing the oxides and the solutions. The weighings of the oxides and solutions were performed using the substitution weighing technique. The certified mass values obtained in 2009 are traceable to the SI via the International Kilogram Prototype and regular calibrations of the JRC-Geel principle kilogram. The relative uncertainty (k=2) from weighing was kept lower than 0.02 %.

2.4 Impurity analysis

In previous experiments, the impurity content was measured on samples of uranium batches of highly enriched ^{235}U and also ^{nat}U in order to evaluate different purification methods. Total impurity levels considerably lower than 100 mg/kg were achieved. Since the starting materials used for the preparation of the IRMM-3000 series were submitted to the same purification cycle, the assumption was made to assign the same level of impurities of 100 mg/kg ± 100 mg/kg (rectangular distribution) to each of the materials. This assumption has been made for the preparation of the IRMM-074 series [4] for the enriched ^{235}U starting material BC02154 in the combination with the previously used ^{238}U starting material BC02155. The latter was exhausted at the time of the IRMM-3000 preparation in 2009, therefore another ^{238}U material, the BC02677, had to be used for the IRMM-3000 series.

The validity of the assumed equal impurity levels for the starting materials used, in this case BC02154 for ^{235}U and BC02155 for ^{238}U, is strongly supported by the excellent agreement of verification measurements for the $n(^{235}U)/n(^{238}U)$ ratio of the IRMM-074 series by TIMS compared to the certification data based on mass metrology. This is shown in Table 1 below [4]. The verification measurements of the $n(^{235}U)/n(^{238}U)$ isotope ratio, the so-called “major” ratio, included the following materials: IRMM-072-1 [7], IRMM-199, NBL U500, IRMM-074-1 and IRMM-074. IRMM-074 represents the "basic mixture" of ^{235}U and ^{238}U, prepared prior to adding ^{233}U later on. This verification measurement series includes some of the worldwide most recognized reference materials with $n(^{235}U)/n(^{238}U)$ ratios close to unity. The verification measurements were performed using the modified total evaporation (MTE) method on the Triton TIMS [8,9,10]. The sample loading was 5 µg of U for all materials. IRMM-072-1 was used as a calibrant, which means the measured $n(^{235}U)/n(^{238}U)$ ratios of IRMM-072-1 were used to determine the K-factor which is then used to perform an external mass fractionation correction on the $n(^{235}U)/n(^{238}U)$ ratios for all other materials.

As shown in Table 1, the results for the $n(^{235}U)/n(^{238}U)$ ratios for IRMM-199 and NBL U500 agree well with the certified values, showing that the employed method provides accurate data. Remarkably, the $n(^{235}U)/n(^{238}U)$ ratio of NBL U500 shows a relative difference of 0.04 %. This difference is not significant because of the 0.11 % uncertainty, which is mainly arising from the 0.1 % uncertainty of the certified value. The relative difference of about 0.04 % was confirmed by Cheng et al. [11] using IRMM-074-1 for mass fractionation correction. Furthermore the corrected $n(^{235}U)/n(^{238}U)$ ratios for IRMM-074-1 and IRMM-074 agree well at a level of 0.02 % for the (insignificant) relative difference to the certified values obtained from the mixture calculation. This confirms that the assumption regarding the equal impurities for the starting materials BC02154 and BC02155 was valid. The verification measurements for the $n(^{233}U)/n(^{235}U)$ ratios for the IRMM-074 series were successful at a similar level as well [4], which confirms that the assumption regarding the equal impurities even applies to all used starting materials of the IRMM-074 series, which includes the ^{233}U starting material BC02153.

Table 1. TIMS measurements of the $n(^{235}U)/n(^{238}U)$ isotope ratio for IRMM-072, IRMM-199, NBL U500, IRMM-074-1 and IRMM-074, using the modified total evaporation technique [8,9]. IRMM-072-1 was used as calibrant (* no result given) for the mass fractionation correction. Expanded uncertainties with a coverage factor k=2 are given in parentheses and apply to the last 2 digits of the measurement value.

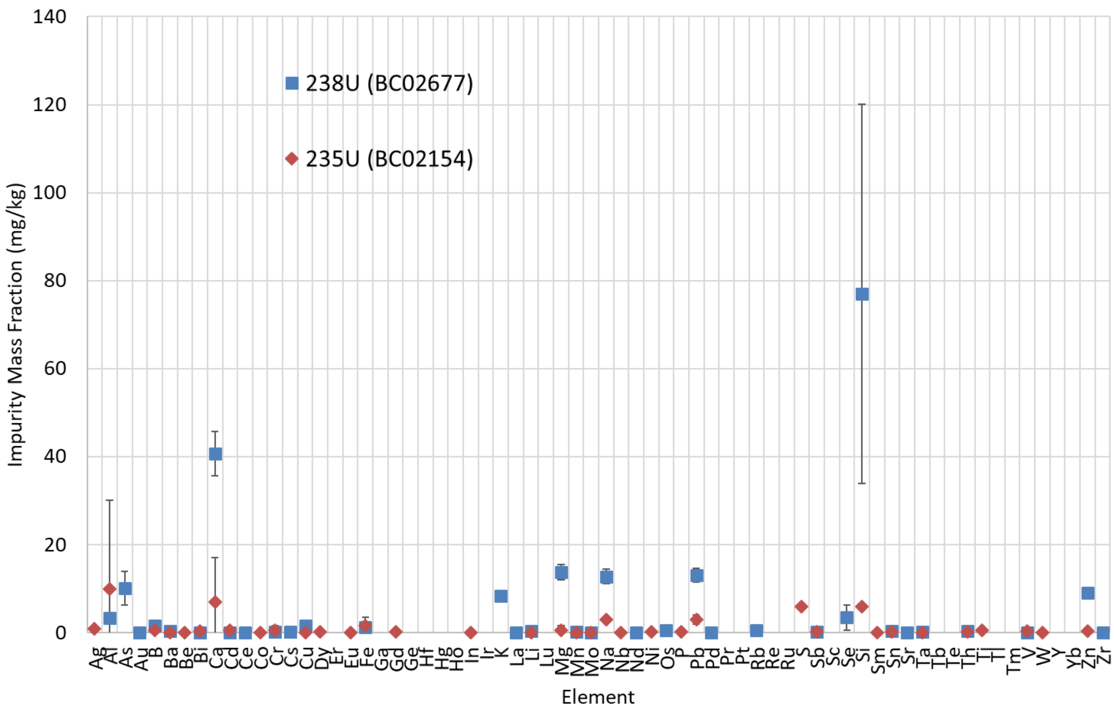
	Measured $n(^{235}U)/n(^{238}U)$ (and corrected using IRMM-072-1)	Certified $n(^{235}U)/n(^{238}U)$	Rel. Dev from certified value
IRMM-072-1	*	0.99103(20)	N/A
NBL U500	1.00009(37)	0.9997(10)	0.04(11)%
IRMM-199	1.00018(36)	1.00015(20)	0.003(42)%
IRMM-074	1.00009(43)	1.00026(15)	-0.017(44)%
IRMM-074-1	1.00022(32)	1.00025(15)	-0.003(33)%

The assumption regarding equal impurities in the starting U materials has also been shown at a level of 0.02% accuracy by verification measurements for the $n(^{236}U)/n(^{238}U)$ ratios of the IRMM-075 series [5,12]. This is a gravimetrically prepared mixture between a ^{236}U spike (IRMM-3660) and a natural uranium material, which have been purified and calcinated together using the same procedure as for the IRMM-074 series.

Furthermore, a double spike IRMM-3636a [6] with a $n(^{233}U)/n(^{236}U)$ ratio close to unity has been prepared in the same way. The verification measurements for IRMM-3636a were performed using a high precision double spike method, which is characterized by an internal rather than external (as for MTE) mass fractionation correction using the $n(^{235}U)/n(^{238}U)$ ratio of IRMM-074. The results show that the difference of the measured $n(^{233}U)/n(^{236}U)$ ratio to the certified value obtained from the gravimetrical calculation was at a level of 0.003(16) %. This excellent agreement confirms the assumptions of achieving similar and correlated levels for impurity and stoichiometry for all enriched starting materials of ^{233}U, ^{235}U, ^{236}U and ^{238}U and the natural material, used for the preparation of the various gravimetrical mixtures in 2004.

However, the observations during the verification measurements for the IRMM-3000 series indicate the presence of a possibly higher impurity content for the ^{238}U material BC02677, which might originate from the source of the material itself or was not sufficiently removed during the purification procedure performed several years later (in 2009). Therefore additional impurity measurements for the ^{238}U oxide starting material BC02677 have been initiated and performed in 2019 by ICPMS at the Analytical Services of the JRC-G.II.8 unit in Karlsruhe/Germany. In Figure 1 the results are compared to the impurity measurements for the ^{235}U oxide starting material BC02154 performed in 2005 by ICPMS at the SCK/CEN in Mol/Belgium.

Figure 1. Impurity Mass Fractions for the ^{238}U (BC02677) and ^{235}U (BC02154) Starting Materials (Uncertainties: k = 2).



The total impurity mass fraction, *i.e.* the sum of all elemental impurity mass fractions for the ^{238}U material BC02677, was determined to be 199 mg/kg ± 89 mg/kg. The uncertainty (with a coverage factor k=2) is calculated using the uncertainties of all quantified impurities with a mass fraction value higher than zero, and also by taking into account the given "limits of quantification" (considered as 99 % confidence level and k = 3) for elements for which a quantification was not possible. The most abundant impurity elements for the ^{238}U material are Si (77 mg/kg ± mg/kg 43 mg/kg) and Ca (40 mg/kg ± 5 mg/kg), followed by As, Mg, Na and Pb with mass fractions between 10 mg/kg and 20 mg/kg. The large (>50 % relative) uncertainty for Si is due to Argon related interferences occurring within the ICPMS instrument. Earlier impurity analyses for the ^{238}U starting material performed in 2010 at other laboratories did not report any results for the most abundant impurity element Si, most likely due to these interferences causing Si to be quite difficult to measure. This led to an initial underestimation of the total impurity mass fraction of the ^{238}U material BC02677, and might explain some of the earlier observed relative differences between the verification measurements by TIMS and the ratios calculated from the gravimetrical preparation.

As shown in Figure 1, the impurity mass fractions for the ^{235}U material BC02154 are much lower compared to the ^{238}U material BC02677, also for the major impurity elements. Re-calculating the total impurity mass fraction with its uncertainty for the ^{235}U material BC02154 in the same manner as for the ^{238}U material leads to a result of 42 mg/kg ± 23 mg/kg (with a coverage factor k=2). This result was subsequently used for an update of the gravimetrical calculations of isotope ratios of the IRMM-3000 series, instead of the previously used value of 100 mg/kg ± 100 mg/kg

In contrast to the assumption of similar and correlated impurities for the IRMM-074 and IRMM-075 series and the double spike IRMM-3636a, which were justified by the respective verifications measurements, for the IRMM-3000 series the impurities for the used ^{235}U and ^{238}U starting materials are obviously different and cannot be considered as correlated.

2.5 Determination of the isotopic ratios in starting materials

The isotopic compositions of the ^{235}U and ^{238}U starting materials of the IRMM-3000 series were measured in 2009 using the MTE method on the Triton type TIMS (Thermo Scientific, Bremen, Germany) [8,9,10]. The results are shown in Table 2 and Table 3, respectively. The TIMS instrument was calibrated using the IRMM-184 isotope reference material (see certificate in Annex 1). The ^{235}U starting material BC02154 was already measured within the IRMM-074 certification project [4] in 2004. In order to avoid mutual background contributions the ^{235}U and ^{238}U starting materials were measured on different cleaned sample turrets and in different measurement sequences. The reference material IRMM-184 was used for mass fractionation correction with a close to natural isotopic composition, and was not measured on the same turret as the ^{235}U starting material BC02154. Instead, the IRMM-184 was measured on the same turret as the previous ^{238}U starting material BC02155, which was exhausted at the end of the IRMM-074 project. The uncertainty contribution from this external type of mass fractionation correction consitutes only a minor component of the uncertainty for the corrected ratios of BC02154. The new ^{238}U starting material BC02677 for the IRMM-3000 series was measured separately in 2009, and calibrated again using IRMM-184, in this case being measured on the same turret.

Table 2. Isotopic composition of highly enriched ^{235}U starting material BC02154.

	Certified amount ratios		
$n(^{234}U)/n(^{235}U)$	0.000021283(87)		
$n(^{236}U)/n(^{235}U)$	0.00004228(19)		
$n(^{238}U)/n(^{235}U)$	0.0000010239(27)		
	Certified molar mass		
M_U	235.0439523(24) g/mol		
	Certified amount fractions		Certified mass fractions
$n(^{234}U)/n(U)$	0.000021282(87)	$m(^{234}U)/m(U)$	0.000021191(87)
$n(^{235}U)/n(U)$	0.99993541(21)	$m(^{235}U)/m(U)$	0.99993531(21)
$n(^{236}U)/n(U)$	0.00004228(19)	$m(^{236}U)/m(U)$	0.00004246(19)
$n(^{238}U)/n(U)$	0.0000010238(27)	$m(^{238}U)/m(U)$	0.0000010369(28)

Table 3. Isotopic composition of highly enriched ^{238}U starting material BC02677.

	Certified amount ratios		
$n(^{234}U)/n(^{238}U)$	0.00000000602(18)		
$n(^{235}U)/n(^{238}U)$	0.000007766(15)		
$n(^{236}U)/n(^{238}U)$	0.00000009344(43)		
	Certified molar mass		
M_U	238.0507634(32) g/mol		
	Certified amount fractions		Certified mass fractions
$n(^{234}U)/n(U)$	0.00000000602(18)	$m(^{234}U)/m(U)$	0.00000000592(18)
$n(^{235}U)/n(U)$	0.000007766(15)	$m(^{235}U)/m(U)$	0.000007668(14)
$n(^{236}U)/n(U)$	0.00000009341(43)	$m(^{236}U)/m(U)$	0.00000009266(42)
$n(^{238}U)/n(U)$	0.999992134(15)	$m(^{238}U)/m(U)$	0.999992233(15)

2.6 Preparation of mixtures and solutions

The IRMM-3000 series was produced in 2009 by carefully weighing of the ^{235}U and ^{238}U oxides and dissolving them in nitric acid. The masses of the starting materials and solutions are shown on the weighing certificate E3731 in Annex 2 and summarized in Table 4. The U mass fraction of the solutions is about 5 mg U /g solution.

Table 4. Masses of starting materials and solutions, and U mass fractions.

	g ^{235}U oxide	g ^{238}U oxide	g solution
IRMM-3020	0.26112(3)	1.03822(5)	253.388(5)
IRMM-3035	0.25812(3)	0.47673(5)	128.574(5)
IRMM-3050	0.29850(3)	0.29481(3)	103.176(5)*
IRMM-3075	0.81046(5)	0.29067(3)	183.194(5)*
IRMM-3090	2.25012(10)	0.34584(3)	423.584(5)

*The verification of the weighing certificate E 3731 with the original records shows agreement of all determined masses except for the masses of the IRMM-3050 and IRMM-3075 solutions, but these are not relevant for the calculated $n(^{235}U)/n(^{238}U)$ ratios, only for the mass fractions of the solutions. In this table for the IRMM-3050 and IRMM-3075 solutions, the results from the original calculations are presented.

2.7 Calculation of the isotope ratios from the mixing of the starting materials

The isotope amount ratios of the individual units and their uncertainties were calculated according to Guide to the Expression of Uncertainty in Measurement [3]. The following contributors to the final uncertainties of the major isotopic ratio $n(^{235}U)/n(^{238}U)$ were recognised during the preparation:

1. Uncertainties from weighing of the starting materials: the expanded relative uncertainties (k=2) range between 0.004 % and 0.012 %. The relative uncertainty contributions from the weighings to the uncertainties of the isotope ratios of the mixtures are significant, ranging between 9.8 % and 56 %.
2. Total impurity mass fractions: based on the levels of impurities for each of the starting materials as explained above. The relative uncertainty contributions from the total impurity mass fractions to the uncertainties of the isotope ratios of the mixtures are significant, ranging between 1.4 % and 41 %.
3. Measurements of the individual isotopic amount ratios for the ^{235}U and ^{238}U starting materials. The uncertainties are negligible for the calculation of the isotope ratios in the mixtures.
4. Stoichiometry: the ^{235}U and ^{238}U starting materials were calcinated at the same temperature and humidity conditions, which is expected to lead to the same stoichiometry. For the U_3O_8 oxide a value of 8 was assumed for the oxygen with an uncertainty of 0.01 % with a rectangular distribution indicating the limit values for the stoichiometry. A correlation factor of 1.0 was applied for the stoichiometry (i.e. the oxygen value of 8) for the ^{235}U and ^{238}U starting materials due to the calcination under identical conditions. As a consequence, the uncertainty contributions from the stoichiometry to the uncertainties of the isotope ratios of the mixtures are therefore equal to zero.

The calculations from the mixing procedure of the isotope ratios for the five mixtures of the IRMM-3000 series for $n(^{235}U)/n(^{238}U)$ and $n(^{238})/n(^{235}U)$ are shown in Annex 3 for IRMM-3020 as an example. For the other materials the calculations are similar except for the entries of the masses for the ^{235}U and ^{238}U oxide starting materials as given in Table 4. The results for the isotope ratios are shown in Table 5.

Table 5. Calculated isotope ratios for the IRMM-3000 series. All uncertainties are given with coverage factor k=2.

$n(^{m}U)/n(^{238}U)$	$n(^{235}U)/n(^{238}U)$	$n(^{238}U)/n(^{235}U)$
IRMM-3020	0.254264(41)	3.93292(63)
IRMM-3035	0.54736(10)	1.82694(34)
IRMM-3050	1.02359(18)	0.97696(17)
IRMM-3075	2.81872(44)	0.354771(56)
IRMM-3090	6.57731(92)	0.152038(21)

The measurement of the so-called “minor” isotope ratios $n(^{234})/n(^{235}U)$, $n(^{236})/n(^{235}U)$, $n(^{234})/n(^{238})$, $n(^{236})/n(^{238}U)$ in the highly enriched ^{235}U and ^{238}U starting materials was compromised by the peak tailing from the major isotope beams, therefore the calculated minor ratios of the mixtures are not very reliable and not usable for the certification. Instead of using the calculated ratios from the starting materials, the minor ratios were directly characterized on the IRMM-3000 series solutions and certified using TIMS/MTE measurements in 2019 at JRC-G.2, and were subsequently verified by laboratories at the IAEA and ORNL.

The uncertainties for the major isotope ratios $n(^{235}U)/n(^{238}U)$ (or expressed as $n(^{238}U)/n(^{235}U)$) range between 0.014 % and 0.019 % as shown in the uncertainty budgets in Table 6. The masses of the ^{235}U and ^{238}U oxide starting materials dominate the uncertainty budgets and their relative contributions clearly depend on the mixing proportions as represented as well by the isotope ratios. The relative uncertainty contributions from the impurities of the ^{238}U starting material are consistently higher compared to those from the ^{235}U starting material, they both depend on the mixing proportions and ratios as well.

Table 6. Uncertainty budgets for the "major" ratios $n(^{235}U)/n(^{238}U)$ for the IRMM-3000 series, calculated from the gravimetrical mixing (*due to the rounding of the last digit the sum of the relative uncertainty contributions can deviate by 0.1 % from the expected 100 %). As explained below, in case of IRMM-3035, the "major" ratio $n(^{235}U)/n(^{238}U)$ calculated from the gravimetrical mixing is not used for the certification.

	IRMM-3020	**IRMM-3035**	**IRMM-3050**	**IRMM-3075**	**IRMM-3090**
Ratio $n(^{235}U)/n(^{238}U)$	0.254264(41)	0.54736(10)	1.02359(18)	2.81872(44)	6.57731(92)
Rel. Unc. (k=2), in %	0.016	0.019	0.017	0.016	0.014
Relative uncertainty contributions in %(*)					
Mass ^{235}U oxide	51.4	39.0	33.0	15.5	10.1
Mass ^{238}U oxide	9.0	31.8	33.8	43.3	38.3
Impurities ^{235}U	2.1	1.5	1.7	2.2	2.7
Impurities ^{238}U	37.4	27.7	31.4	39.1	48.9

2.8 The Dispensing of the IRMM-3000 Series

Dispensing of the IRMM-3000 solutions with a mass fraction of about 5 mg U /g into quartz ampoules and the flame sealing was carried out in a double section fume hood in the controlled area at JRC-Geel. The ampoules were filled with 0.5, 1 and 5 mL of solution by means of a dispenser.

Table 7. Sequence of ampoule filling for the IRMM-3000 series in 2009. Several units have been used at JRC-G.2 in Geel for analyses and several more units were transferred to laboratories at the IAEA, JRC-G.II.8 and ORNL for verification analyses.

	Date	Volume	Number of ampoules sealed
IRMM-3020	12 oct 2009	0.5 mL	4
IRMM-3020	12 oct 2009	1 mL	119
IRMM-3020	12 oct 2009	5 mL	32
IRMM-3035	13 oct 2009	1 mL	118
IRMM-3050	13 oct 2009	1 mL	94
IRMM-3075	13 oct 2009	1 mL	118
IRMM-3075	13 oct 2009	5 mL	11
IRMM-3090	14 oct 2009	1 mL	116
IRMM-3090	14 oct 2009	5 mL	55

2.9 Verification Measurements for the $n(^{235}U)/n(^{238}U)$ ratio by Mass Spectrometry

Verification measurements were performed in 2019 by several methods of isotope mass spectrometry. In particular, for the so-called “major” isotope ratio, isotope ratio measurements were performed at JRC-G.2 using the double spike (DS) and modified total evaporation (MTE) methods. This was possible due to the relatively low ^{236}U abundances. For TIMS/DS measurements the double spike reference material IRMM-3636a (see certificate in Annex 1) was used, which is a mixture of highly enriched ^{233}U and ^{236}U starting materials with a ratio of $n(^{233}U)/n(^{236}U) \approx 1$. This method allows an internal mass fractionation correction and therefore leads to lower measurement uncertainties [6]. The DS method was standardized by ASTM-International as C1871-18a in 2018 [13]. For quality control (QC) of the DS method the certified reference materials IRMM-184 (see certificate in Annex 1) and IRMM-074 (see certificate in Annex 1) are used.

Furthermore, and also in order to determine the minor isotope amount ratios with smallest possible measurement uncertainties (see results below), the MTE method was used for verification measurements for the major ratios $n(^{235}U)/n(^{238}U)$ of the IRMM-3000 series. The gravimetrically prepared IRMM-074 isotope reference material was used as calibrant. The MTE method was introduced in 2003 [8,9] and standardized in 2016 by ASTM-International as C1832-16 [10].

The TIMS analyses for all 5 materials and for all methods were performed on loading solutions taken from the equivalent of 2 ampoules of 5 mL each, with at least 3 replicate filament loadings. There was no case of disagreement between the results of the 2 loading solutions, therefore the results for each of the 5 materials and for each of the used methods were combined.

Figure 2. Relative differences of TIMS measurements of the major ratios $n(^{235}U)/n(^{238}U)$ to the gravimetrically calculated ratios.

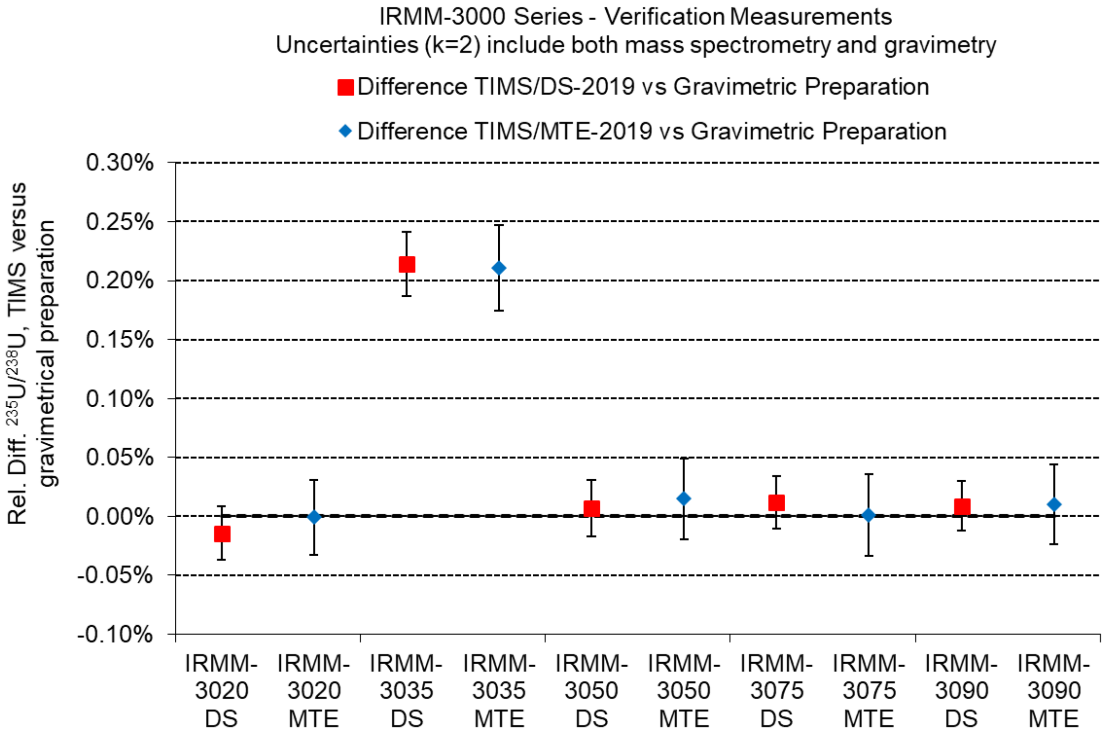
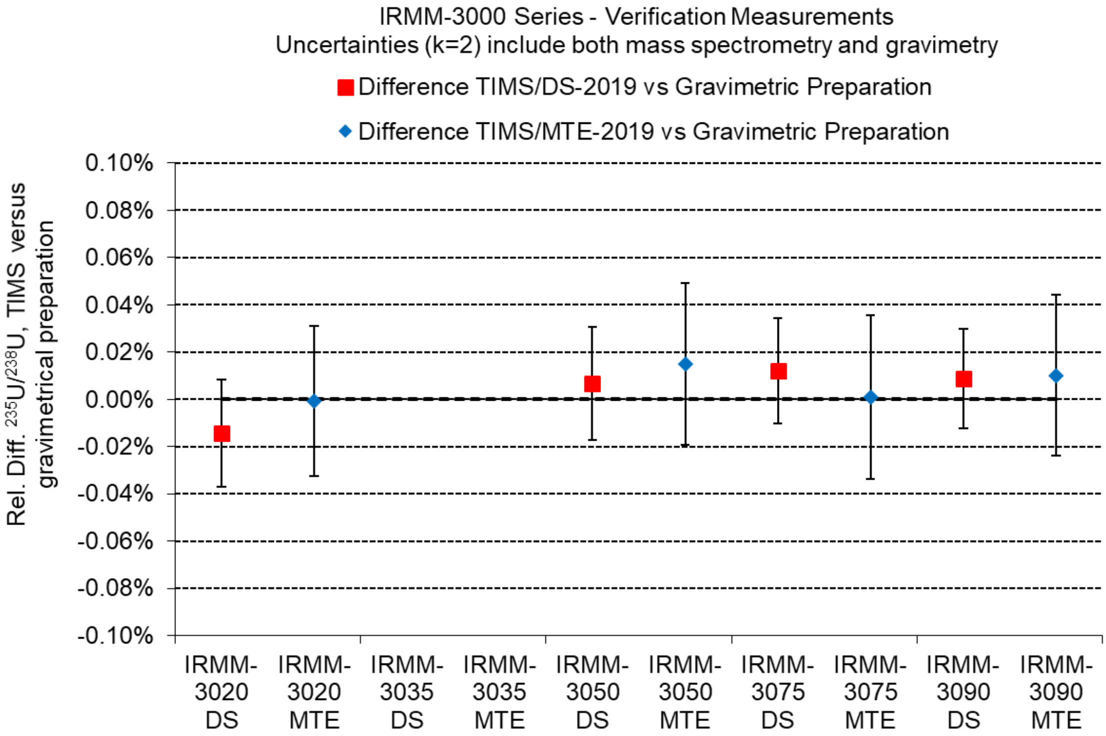


Figure 3. Closer focus on the relative differences of TIMS measurements of the major ratios $n(^{235}U)/n(^{238}U)$ to the gravimetrically calculated ratios (IRMM-3035 is out of scale).



As shown in Figure 2 and Figure 3, the gravimetrically calculated $n(^{235}U)/n(^{238}U)$ ratios for IRMM-3020, IRMM-3050, IRMM-3075 and IRMM-3090 agree well with the mass spectrometric results obtained using the TIMS/DS and TIMS/MTE methods. Therefore, the gravimetrically calculated ratios are confirmed by an independent instrumental technique such as TIMS using two different methods such as TIMS/DS and TIMS/MTE and at a level for the accuracies and uncertainties of about 0.02%. As a consequence, the $n(^{235}U)/n(^{238}U)$ ratios from the gravimetric preparations for IRMM-3020, IRMM-3050, IRMM-3075 and IRMM-3090 can be used for the certification in compliance with ISO 17034 [1].

In contrast, the $n(^{235}U)/n(^{238}U)$ ratio for IRMM-3035 was not confirmed by the mass spectrometric measurements. The reason for this is unknown. The weighing conditions such as the relative humidity during the weighing of the IRMM-3035 starting materials were as good (about 47 %-48 %) as during the weighing of all the other materials of the series. The weighing of the ^{235}U and ^{238}U starting materials for IRMM-3035 was performed on the same day (28/09/2009) as the weighing for the ^{235}U and ^{238}U starting materials for IRMM-3050 and for the ^{235}U staring material for IRMM-3075, followed on the next day by the ^{238}U starting material for IRMM-3075 and both starting materials for IRMM-3090. The weighings for IRMM-3020 were performed 3 days before those of IRMM-3035. There is no explanation for the relative difference of about 0.21 % of the measured $n(^{235}U)/n(^{238}U)$ ratio from the gravimetrically calculated one. But this range of deviation is far beyond the performances, i.e. accuracies and uncertainties, known for both the applied mass metrological and mass spectrometric techniques, and it is therefore not acceptable for using the gravimetrically calculated ratio for the certification.

However, the $n(^{235}U)/n(^{238}U)$ ratio for IRMM-3035 measured by the TIMS/DS method was confirmed by TIMS/MTE measurements at JRC-Geel. The measured $n(^{235}U)/n(^{238}U)$ ratio for IRMM-3035 was furthermore confirmed by measurements at the IAEA-SGAS and at ORNL using TIMS/MTE and MC-ICPMS methods, as shown in Table 8. The confirmation measurements show insignificant differences, which are fully covered by the measurement uncertainties. The result of 0.03(11) % from ORNL by TIMS/MTE is normalized to NBL U500. If this is re-normalized to IRMM-2025 (a certified uranium solution with about 2 % enrichment of ^{235}U), which was measured as QC sample by ORNL, the result for the relative difference would be 0.001(47) %. This overall good agreement allows the certification of the $n(^{235}U)/n(^{238}U)$ ratio for IRMM-3035 using the result of the high precision TIMS/DS method obtained at JRC-Geel.

Table 8. Mass spectrometric results for the $n(^{235}U)/n(^{238}U)$ ratio for IRMM-3035. The uncertainties are given with coverage factor k=2. (NML= Nuclear Material Laboratory, ESL= Environmental Sample Laboratory)

Laboratory and Method	$n(^{235}U)/n(^{238}U)$	Rel. Diff. to JRC-Geel, TIMS/DS
JRC-Geel, TIMS/DS	0.548531(89)	
JRC-Geel, TIMS/MTE	0.54851(16)	-0.003(29) % (vs. IRMM-074)
IAEA/NML, TIMS/MTE	0.54846(20)	-0.012(37) % (vs. IRMM-184)
IAEA/ESL, MC-ICPMS	0.54853(19)	-0.001(35) % (vs. IRMM-2025)
ORNL, TIMS/MTE	0.54839(60)	-0.03(11) % (vs. NBL U500)
ORNL, MC-ICPMS	0.54866(21)	+0.023(41) % (vs. IRMM-074)

2.10 Characterization of the isotope ratios by TIMS/MTE combined with gravimetry and TIMS/DS

The material characterisation is the process of determining the property value(s) of a reference material. The material characterisation for the IRMM-3000 series was based on a primary direct method (gravimetry) [14] and confirmed by independent isotope ratio measurement methods such as TIMS/DS and TIMS/MTE, also by external collaborators. A primary method of measurement (also called "primary reference method" in the International Vocabulary of Metrology (VIM) [15]) is a method that does not require calibration with a standard of the same measurand and does not depend on a chemical reaction[1]. Such methods are of highest metrological order and often yield results with low uncertainties. However, it is nevertheless prudent to demonstrate absence of bias or gross errors by use of an independent method of lower metrological order.

The characterization of the $n(^{235}U)/n(^{238}U)$ major ratios was obtained by gravimetrical calculations, except for IRMM-3035 for which the confirmed TIMS/DS ratio was used, as described in sections 0 and 2.9 above. For the characterization of the minor ratios, the results from the TIMS/MTE measurements performed in 2019 at JRC-Geel according to [8,9,10] were used. The MTE method has been recently upgraded at JRC-Geel for the use of Faraday cup amplifiers with 10^{13} Ω resistors in order to reduce the signal-to-noise ratio for measurement of the minor isotopes ^{234}U and ^{236}U, leading to lower uncertainties for the minor ratios $n(^{234}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$ [12]. The upgrade of the method requires longer integration and waiting times for the measurement of the minor isotopes and a better control of the instrument mass calibration. The details of the updated MTE method and QC measurement results using the IRMM-075 series are described in [12] and in the draft revised version of the ASTM C1832-21 standard document. The results of the gravimetrical data and the double spike method [6,13] for the $n(^{235}U)/n(^{238}U)$ major ratios provide an advantage for the internal mass fractionation correction of the minor ratios acquired by the MTE method, instead of using the major ratio data acquired by the MTE method itself which have slightly higher uncertainties. The results of the characterization are shown in Table 9 and Annex 4-Annex 8.

Table 9. Characterization of the isotope ratios for the IRMM-3000 series. The minor ratios are normalized using the characterized major ratios $n(^{235}U)/n(^{238}U)$ obtained by gravimetrical calculations, except for IRMM-3035 (*) for which the confirmed TIMS/DS ratio was used. The uncertainties are given with coverage factor k=2.

	$n(^{234}U)/n(^{238}U)$	$n(^{235}U)/n(^{238}U)$	$n(^{236}U)/n(^{238}U)$
IRMM-3020	0.000004931(35)	0.254264(41)	0.000009805(63)
IRMM-3035	0.000010699(69)	0.548531(89) (*)	0.000021214(86)
IRMM-3050	0.00001991(13)	1.02359(18)	0.00003969(12)
IRMM-3075	0.00005473(35)	2.81872(44)	0.00010966(20)
IRMM-3090	0.00012770(83)	6.57731(92)	0.00025642(34)

[1] *A primary method is "a method having the highest metrological qualities, whose operation(s) can be completely described and understood and for which a complete uncertainty statement can be written in terms of SI units. A primary ratio method measures the value of a ratio of an unknown to a standard of the same quantity; its operation must be completely described by a measurement equation*

2.11 Homogeneity Assessment

A key requirement for any reference material aliquoted into units is equivalence between those units. In this respect, it is relevant whether the variation between units is significant compared to the uncertainty of the certified value, but it is not relevant if this variation between units is significant compared to the analytical variation. Consequently, ISO 17034 [1] requires reference material (RM) producers to quantify the between-unit variation. This aspect is covered in between-unit homogeneity studies.

A homogeneity assessment of these highly enriched uranium materials is not considered to be necessary due to the homogeneous nature of uranium nitrate solutions. There is evidence for this from the certification of the IRMM-019-029 and IRMM-2019-2029 series, where the uncertainty contribution from the homogeneity testing was demonstrated to be at the level of measurement statistics, following the expected trends with the values of the ratios [16,17,18].

2.12 Stability Assessment

2.12.1 Short-term stability study

The IRMM-3000 series consists of uranium nitrate solution isotope reference materials in 1 mol/L HNO_3 contained in flame sealed quartz ampoules. Since the isotopic composition is independent of the temperature, there is no impact from transportation on the uranium isotopic composition. Therefore no short-term stability study was performed and the materials can be dispatched without further precautions under ambient conditions.

2.12.2 Long-term stability study

Long-term stability assessment is not considered to be necessary, due to the stable nature of the uranium isotope ratios in uranium nitrate solutions. This has been demonstrated during the certification of the IRMM-019-029 and IRMM-2019-2029 series, where no significant trends were observed for isotope ratios over a time period of 10 years. Therefore the validity of the certificate is given for 10 years [17,18].

2.13 Uncertainty budgets for minor ratios $n(^{234}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$

Since the materials are homogeneous and stable with time, as demonstrated by experience (as described in sections 2.11 and 2.12), no additional uncertainties other than those already considered for the characterization have to be taken into account for the certification of the isotope ratios. Therefore the uncertainties from the characterization as shown in Table 9 can be used also for the certification. In Table 10 and Table 11 the uncertainty budgets for the "minor" ratios $n(^{234}U)/n(^{238}U)$ and $n(^{234}U)/n(^{238}U)$ are presented.

The measurement of minor ratios for HEU materials like the IRMM-3000 series is more challenging compared to the measurement of LEU materials, because for HEU materials the major isotope beams from ^{235}U and ^{238}U and consequently also their peak tails are a factor of 50-100 higher. Therefore the uncertainties for the minor ratios of the IRMM-3000 series are dominated by the peak tailing effects originated from the ion beams of the major isotopes ^{235}U and ^{238}U towards the minor isotopes ^{234}U and ^{236}U.

The uncertainties for the peak tailing are caused by two effects. Firstly, there are uncertainties of 0.02 amu - 0.03 amu on the mass calibration curve, i.e. the calibration of the detected mass versus the applied magnet current. These cause uncertainties to the tailing-corrected ratios, which depend on the slope of the peak tailing measured at the low and high mass sides of the ion beams. Secondly, there is an uncertainty related to the way of interpolation between the tailing measurements on the low and high mass side of the minor isotope beams. The interpolation can be performed using an arithmetic mean (usually overestimating the tailing contribution) or a geometric mean, which equals the arithmetic mean within the log-scale and takes into account the curvature of the tailing. For the characterization of the IRMM-3000 series the geometric mean was chosen for the tailing correction, in consistency with the verification measurements by the IAEA and ORNL. The uncertainties associated to the peak tailing effects are explained in more detail in Annex 9.

The remaining uncertainties are mainly given by the noise of the Faraday cups, which is minimized due to the use of the $10^{13}\ \Omega$ amplifiers with a better signal-to-noise ratio. But retrospectively, as a consequence of the observed dominating uncertainty from the peak tailing correction, amplifiers with lower resistances, e.g. $10^{12}\ \Omega$ or even $10^{11}\ \Omega$, could have been used as well with only a small increase of the total uncertainty. However, the availability of high precision measurements performed using the $10^{13}\ \Omega$ amplifiers allowed a detailed investigation of the peak tailing effects and the associated uncertainties (see Annex 9).

It appears that the relative uncertainties for minor isotope ratio measurements can be as high as 0.6%, caused by the dominating uncertainty contribution from the peak tailing correction. This is also due to the exceptionally low minor isotope ratios encountered in the IRMM-3000 series, which are HEU materials. However, for HEU isotopic compositions normally encountered in safeguards samples representing typical compositions generated in the nuclear fuel cycle (enrichment, irradiation, reprocessing, re-enrichment) and in HEU reference materials, e.g. like the NBL U500, this uncertainty contribution is smaller and not dominating. For depleted, natural or LEU materials, the uncertainty contribution from the peak tailing correction is usually negligible.

Another contribution to the total uncertainty for the minor ratios of the IRMM-3000 series comes from the normalization using the major ratios. This component is exceptionally small due to the low uncertainties from the gravimetrical preparation, and in case of IRMM-3035 only, the double spike measurement.

Table 10. Uncertainty budgets for the "minor" ratios $n(^{234}U)/n(^{238}U)$ for the IRMM-3000 series. (*due to the rounding of the last digit, the sum of the relative uncertainty contributions can deviate by 0.1 % from the expected 100 %).

	IRMM-3020	**IRMM-3035**	**IRMM-3050**	**IRMM-3075**	**IRMM-3090**
Ratio $n(^{234}U)/n(^{238}U)$	0.000004931	0.000010669	0.00001991	0.00005473	0.00012770
Rel. Unc. (k=2), in %	0.70	0.65	0.64	0.64	0.64
Relative uncertainty contribution in % (*)					
Contribution from measurement of $n(^{234}U)/n(^{238}U)$	18.0	5.1	2.8	1.3	3.7
Major ratio from gravimetry used for normalization	0.1	N/A	0.1	0.1	0.1
IRMM-3636a certified $n(^{233}U)/n(^{236}U)$ ratio	N/A	0.1	N/A	N/A	N/A
Measurement of $n(^{235}U)/n(^{238}U)$ within TIMS/DS	N/A	0.0	N/A	N/A	N/A
Peak tailing	81.9	94.8	97.1	98.6	96.2

Table 11. Uncertainty budgets for the "minor" ratios $n(^{236}U)/n(^{238}U)$ for the IRMM-3000 series (*due to the rounding of the last digit the sum of the relative uncertainty contributions can deviate by 0.1 % from the expected 100 %).

	IRMM-3020	**IRMM-3035**	**IRMM-3050**	**IRMM-3075**	**IRMM-3090**
Ratio $n(^{236}U)/n(^{238}U)$	0.000009805	0.000021224	0.00003969	0.00010966	0.00025632
Rel. Unc. (*k*=2), in %	0.64	0.41	0.31	0.18	0.11
Relative Uncertainty contribution in %(*)					
Contribution from measurement of $n(^{236}U)/n(^{238}U)$	17.9	6.4	12.6	12.1	31.4
Major ratio from gravimetry used for normalization	0.0	N/A	0.1	0.3	0.5
IRMM-3636a certified $n(^{233}U)/n(^{236}U)$ ratio	N/A	0.1	N/A	N/A	N/A
Measurement of $n(^{235}U)/n(^{238}U)$ within TIMS/DS	N/A	0.0	N/A	N/A	N/A
Peak tailing	82.1	93.5	87.3	87.6	68.1

It has to be mentioned, that the use of a SEM detector (secondary electron multiplier) in ion counting mode, in combination with an energy filter (e.g. called "RPQ" on the Triton type TIMS from Thermofisher) instead of Faraday cups for the minor isotopes would allow to reduce the tailing effects and their uncertainties by a factor of 50-100. On the other hand, for this type of measurement, an inter-calibration of the SEM detector versus the Faraday cup detection system has to be performed. As explained in [9,10], this inter-calibration needs to be done preferentially using an ion beam coming from the same sample filament and the same element (uranium). The need for this inter-calibration procedure is firstly given by the fact that the inter-calibration depends on the ion beam focussing within the ion source as well as the "RPQ" energy filter. Secondly, the transmission of the "RPQ" energy filter depends on the initial electrical potential of the ions on the spot where they are created on the filament, and this electrical potential is changing across the length of the filament in order to generate a current through the filament. Even this careful but well-proven type of intercalibration is leading to an additional uncertainty of at least 0.4 % (*k*=2).

Performing the inter-calibration using an ion beam coming from the same sample filament and the same element (uranium), only allowed the inter-calibration to be performed using the ^{235}U ion beam (in case of IRMM-3020, IRMM-3035 and IRMM-3050) or the ^{238}U ion beam of the same sample (in case of IRMM-3050, IRMM-3075 and IRMM-3090), leading to quite low count rates for the minor isotopes ^{234}U and ^{236}U.

In order to investigate the usefulness of this procedure, an additional set of test measurements for the minor isotope ratios of the IRMM-3000 series using the SEM detector for the minor isotopes ^{234}U and ^{236}U was performed in 2021 at JRC-Geel. The results were in agreement with the certified ratios obtained using Faraday cups, but the observed relative uncertainties were at the level of several percent, which is not only due to the inter-calibration but mostly to counting statistics from the measurement of ^{234}U and ^{236}U using the SEM detector in ion counting mode. Therefore, the minor ratio results with smaller uncertainties obtained using Faraday collectors and corrected for the peak tailing (see Table 9) are those used for the certification.

3 Verification Measurements by External Collaborators

Verification measurements for the isotopic composition for the IRMM-3000 series were performed by the IAEA-SGAS/NML using TIMS/MTE and by the IAEA-SGAS/ESL using MC-ICPMS, and additionally by ORNL using TIMS/MTE and MC-ICPMS. In Figure 4 to Figure 18 the results for the verification measurements are presented (full symbols), in combination with quality control (QC) measurements (open symbols) performed for the same ratios. The relative uncertainties for the minor ratios of the IRMM-3000 series are larger compared to the QC materials, because of the larger uncertainty component for the peak tailing correction for HEU materials. For this reason there is also not much difference in the uncertainties depending on the different amplifiers used with 10^{11} Ω or 10^{13} Ω resistors (indicated in the figures as "1E11" and "1E13").

Figure 4. Verification of $n(^{234}U)/n(^{238}U)$ ratio for IRMM-3020 (■) by the IAEA/NML and ORNL, complemented by QC data (□) from the IAEA/NML, ORNL and JRC-Geel. The dashed lines represent the relative uncertainties of the certified ratios. All uncertainties are given with coverage factor k=2.

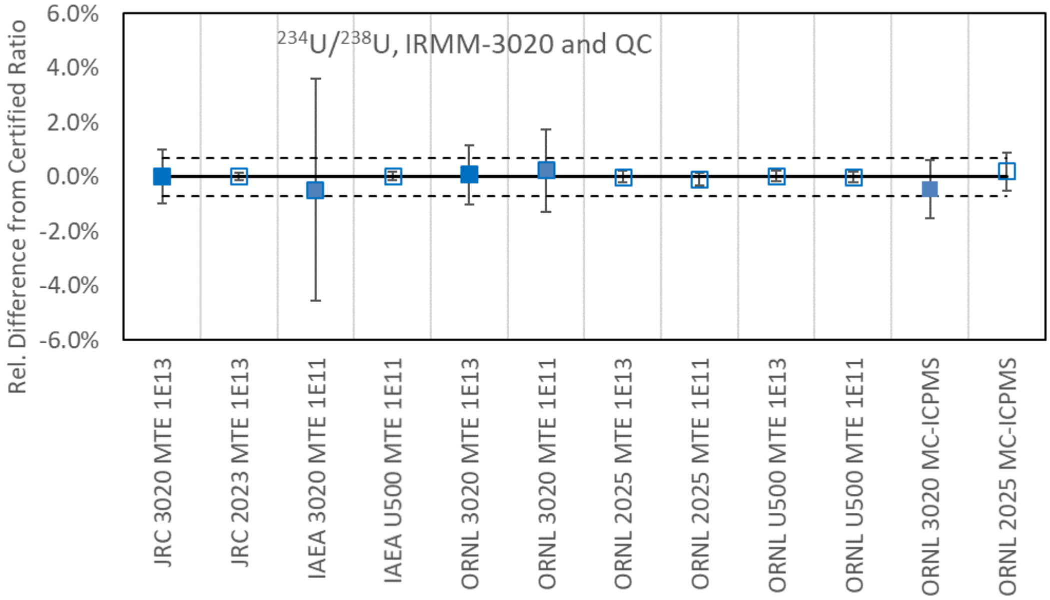


Figure 5. Verification of $n(^{234}U)/n(^{238}U)$ ratio for IRMM-3035 (■) by the IAEA/NML and ORNL, complemented by QC data (□) from the IAEA/NML, ORNL and JRC-Geel. The dashed lines represent the relative uncertainties of the certified ratios. All uncertainties are given with coverage factor k=2.

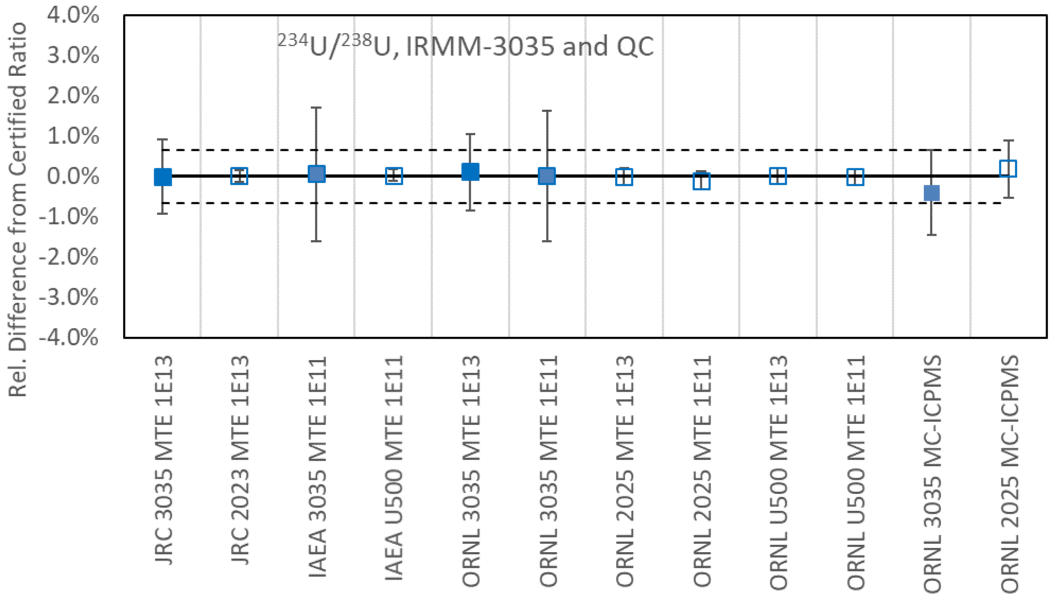


Figure 6. Verification of $n(^{234}U)/n(^{238}U)$ ratio for IRMM-3050 (■) by the IAEA/NML and ORNL, complemented by QC data (□) from the IAEA/NML, ORNL and JRC-Geel. The dashed lines represent the relative uncertainties of the certified ratios. All uncertainties are given with coverage factor k=2.

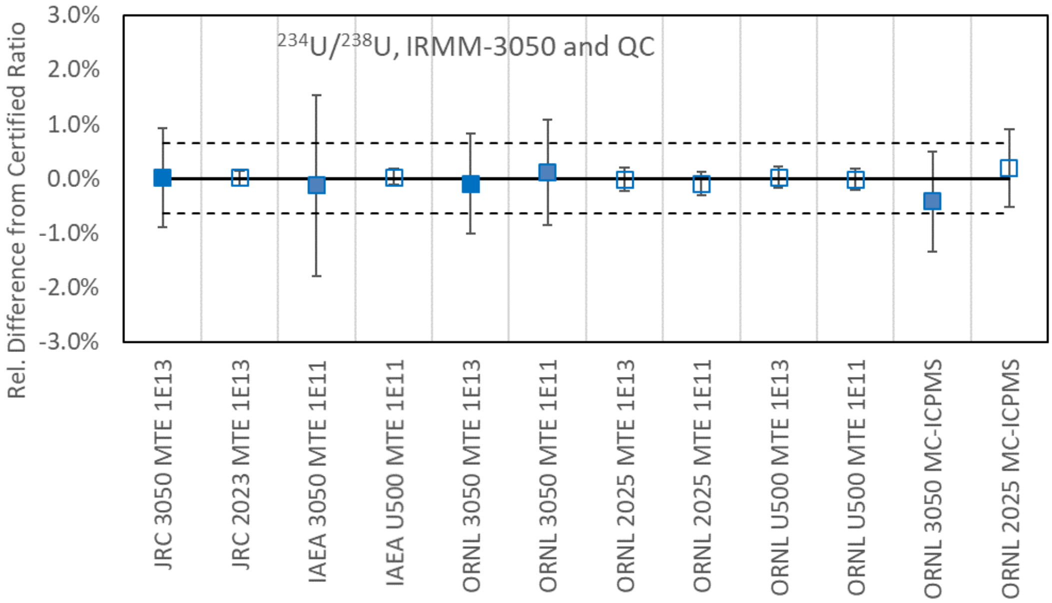


Figure 7. Verification of $n(^{234}U)/n(^{238}U)$ ratio for IRMM-3075 (■) by the IAEA/NML and ORNL, complemented by QC data (□) from the IAEA/NML, ORNL and JRC-Geel. The dashed lines represent the relative uncertainties of the certified ratios. All uncertainties are given with coverage factor k=2.

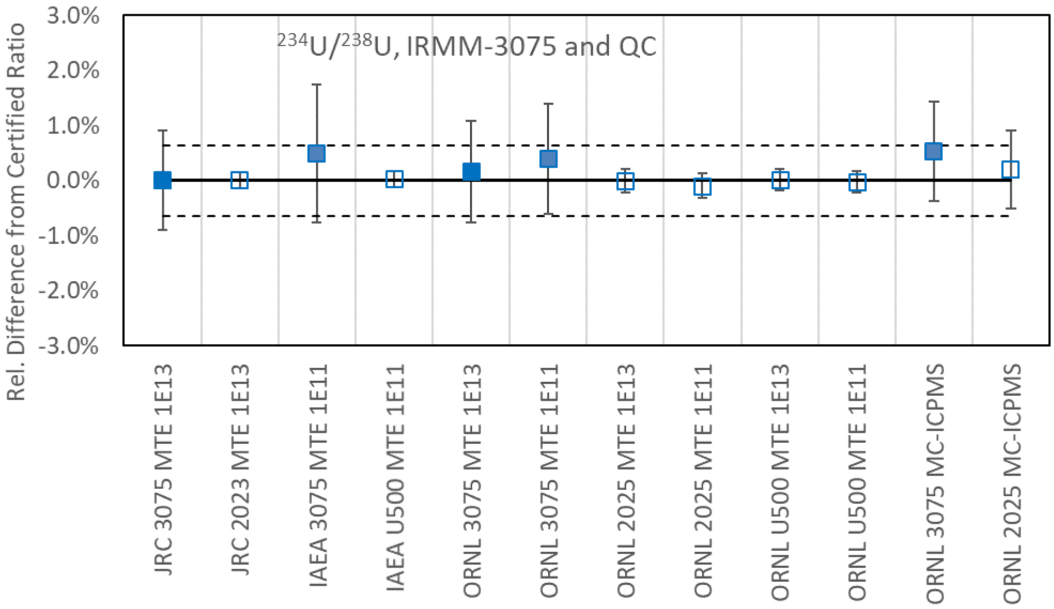


Figure 8. Verification of $n(^{234}U)/n(^{238}U)$ ratio for IRMM-3090 (■) by the IAEA/NML and ORNL, complemented by QC data (□) from the IAEA/NML, ORNL and JRC-Geel. The dashed lines represent the relative uncertainties of the certified ratios. All uncertainties are given with coverage factor k=2.

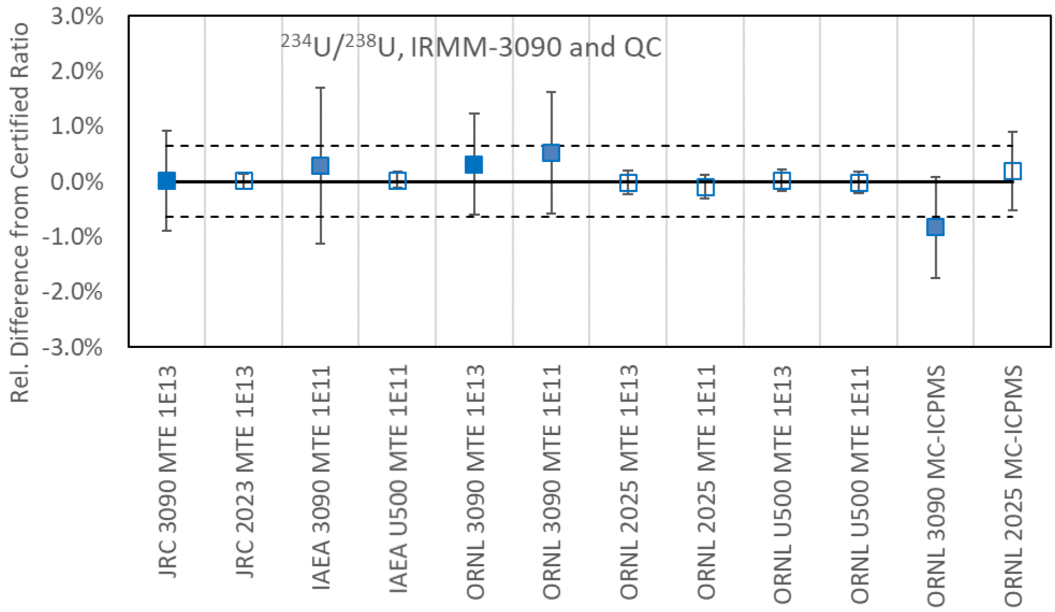


Figure 9. Verification of $n(^{235}U)/n(^{238}U)$ ratio for IRMM-3020 (■) by the IAEA/NML and ORNL, complemented by QC data (□) from the IAEA/NML, ORNL and JRC-Geel. The dashed lines represent the relative uncertainties of the certified ratios. All uncertainties are given with coverage factor k=2.

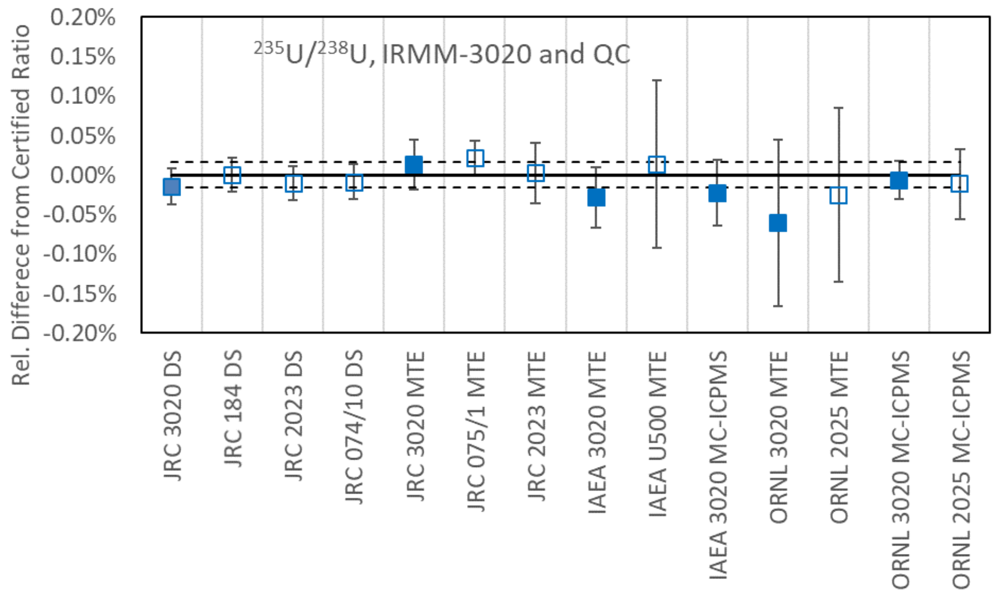


Figure 10. Verification of $n(^{235}U)/n(^{238}U)$ ratio for IRMM-3035 (■) by the IAEA/NML and ORNL, complemented by QC data (□) from the IAEA/NML, ORNL and JRC-Geel. The dashed lines represent the relative uncertainties of the certified ratios. All uncertainties are given with coverage factor k=2.

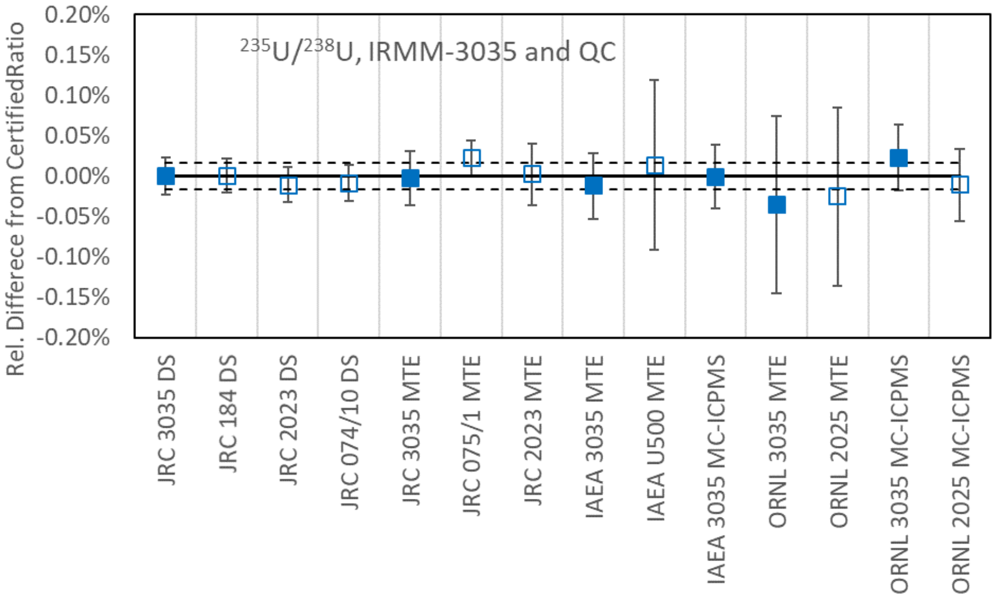


Figure 11. Verification of $n(^{235}U)/n(^{238}U)$ ratio for IRMM-3050 (■) by the IAEA/NML and ORNL, complemented by QC data (□) from the IAEA/NML, ORNL and JRC-Geel. The dashed lines represent the relative uncertainties of the certified ratios. All uncertainties are given with coverage factor k=2.

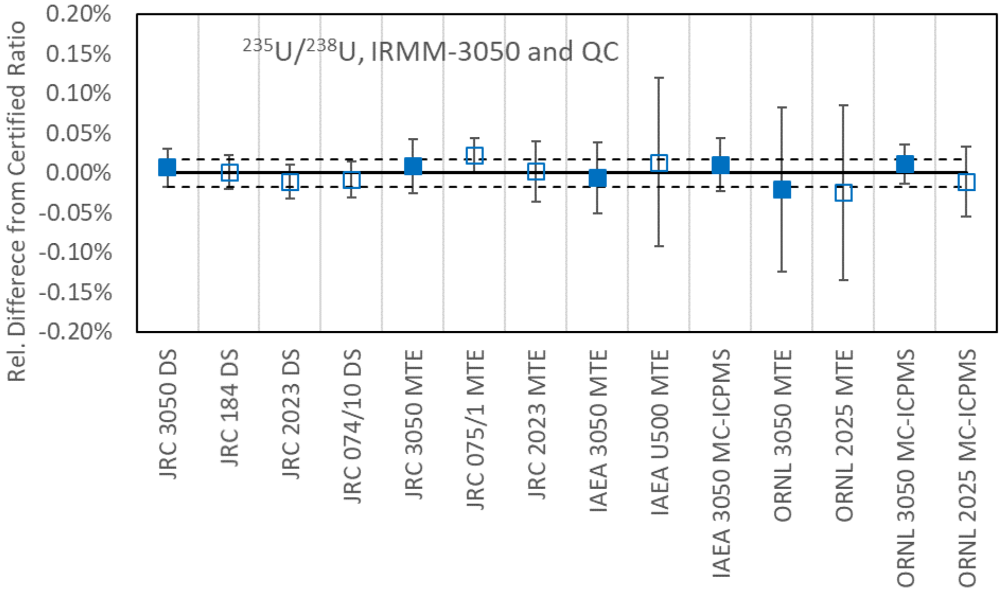


Figure 12. Verification of $n(^{235}U)/n(^{238}U)$ ratio for IRMM-3075 (■) by the IAEA/NML and ORNL, complemented by QC data (□) from the IAEA/NML, ORNL and JRC-Geel. The dashed lines represent the relative uncertainties of the certified ratios. All uncertainties are given with coverage factor k=2.

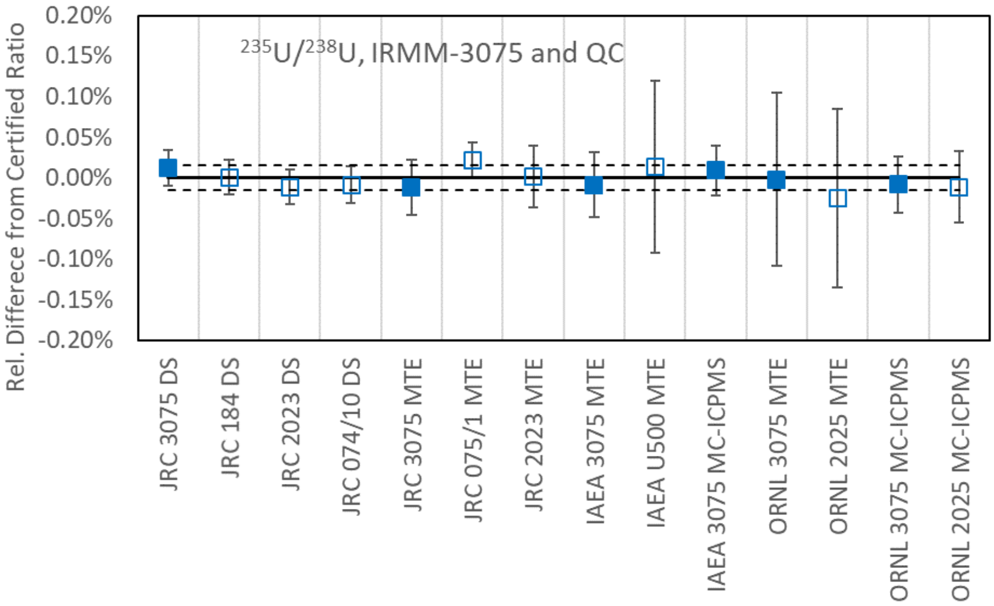


Figure 13. Verification of $n(^{235}U)/n(^{238}U)$ ratio for IRMM-3090 (■) by the IAEA/NML and ORNL, complemented by QC data (□) from the IAEA/NML, ORNL and JRC-Geel. The dashed lines represent the relative uncertainties of the certified ratios. All uncertainties are given with coverage factor k=2.

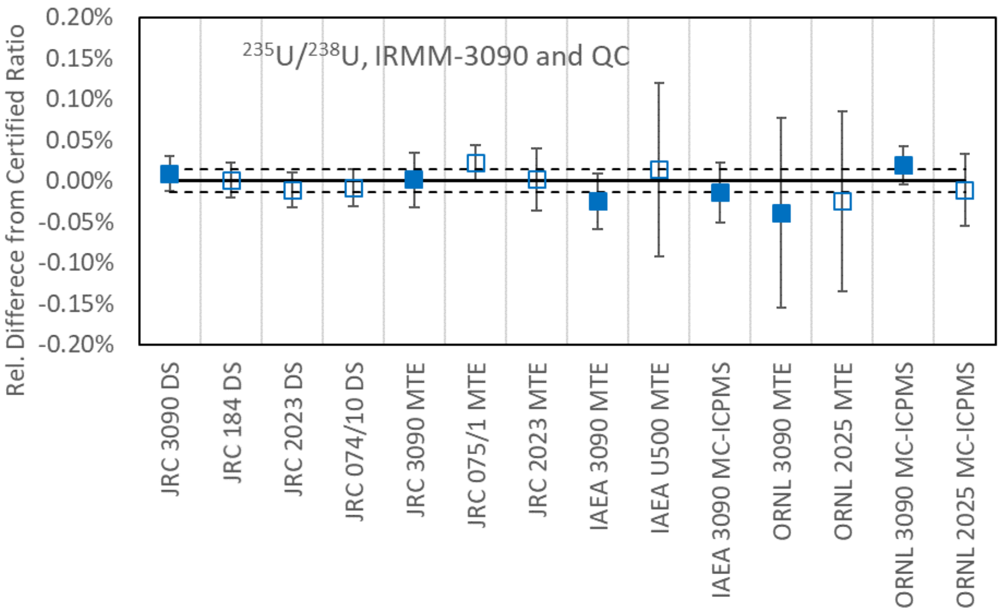


Figure 14. Verification of $n(^{236}U)/n(^{238}U)$ ratio for IRMM-3020 (■) by the IAEA/NML and ORNL, complemented by QC data (□) from the IAEA/NML, ORNL and JRC-Geel. The dashed lines represent the relative uncertainties of the certified ratios. All uncertainties are given with coverage factor k=2.

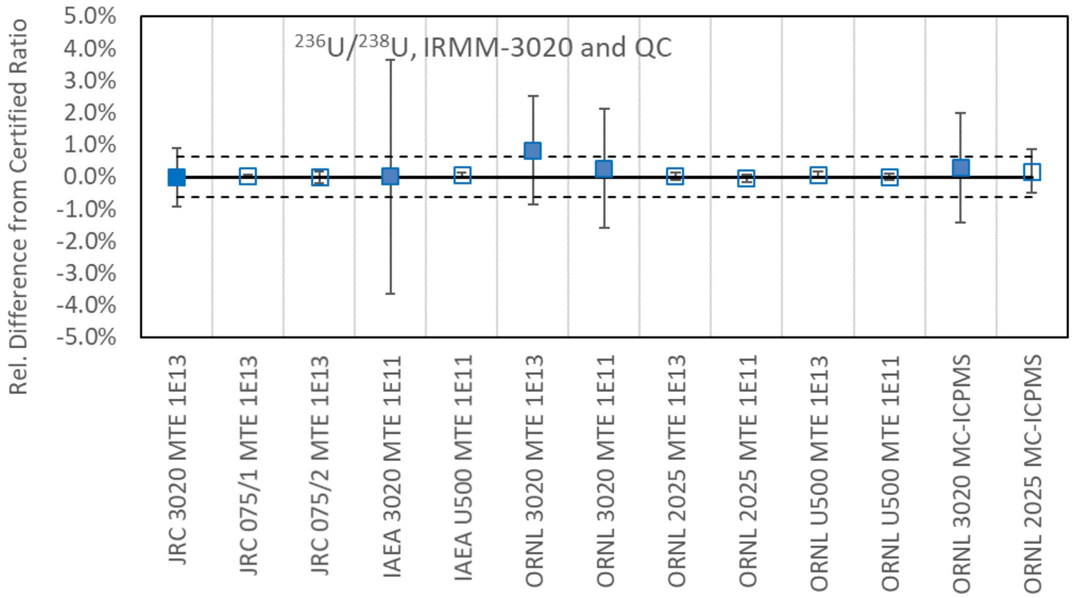


Figure 15. Verification of $n(^{236}U)/n(^{238}U)$ ratio for IRMM-3035 (■) by the IAEA/NML and ORNL, complemented by QC data (□) from the IAEA/NML, ORNL and JRC-Geel. The dashed lines represent the relative uncertainties of the certified ratios. All uncertainties are given with coverage factor k=2.

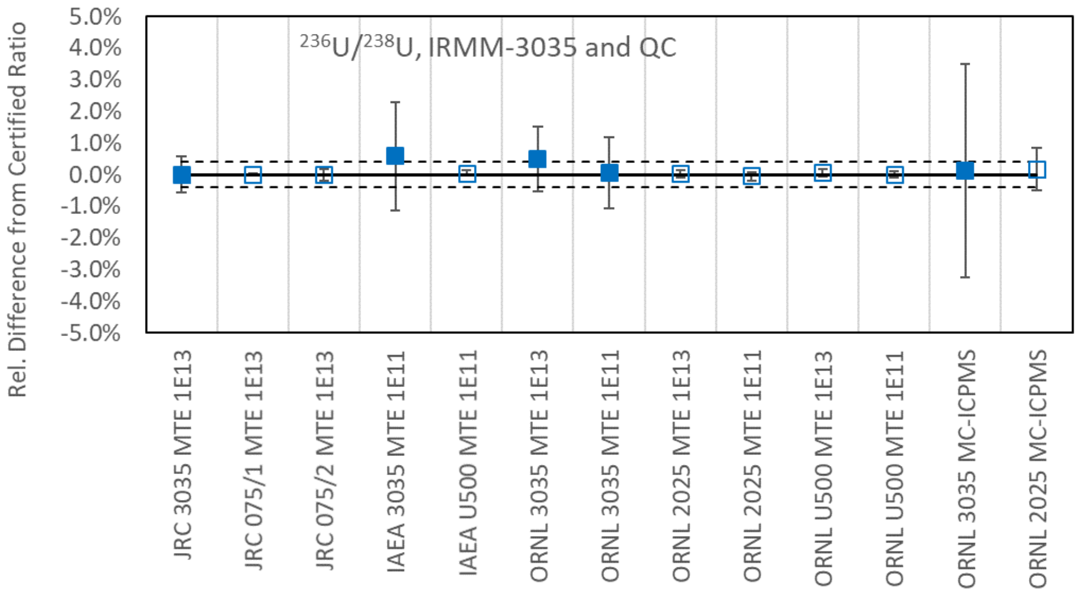


Figure 16. Verification of $n(^{236}U)/n(^{238}U)$ ratio for IRMM-3050 (■) by the IAEA/NML and ORNL, complemented by QC data (□) from the IAEA/NML, ORNL and JRC-Geel. The dashed lines represent the relative uncertainties of the certified ratios. All uncertainties are given with coverage factor k=2.

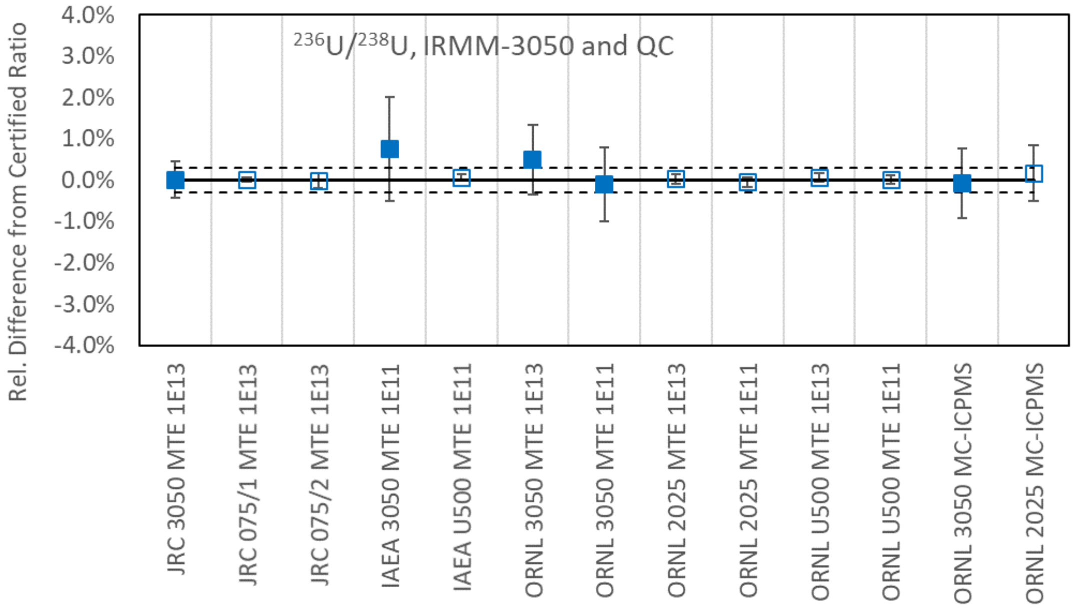


Figure 17. Verification of $n(^{236}U)/n(^{238}U)$ ratio for IRMM-3075 (■) by the IAEA/NML and ORNL, complemented by QC data (□) from the IAEA/NML, ORNL and JRC-Geel. The uncertainties are given with coverage factor k=2.

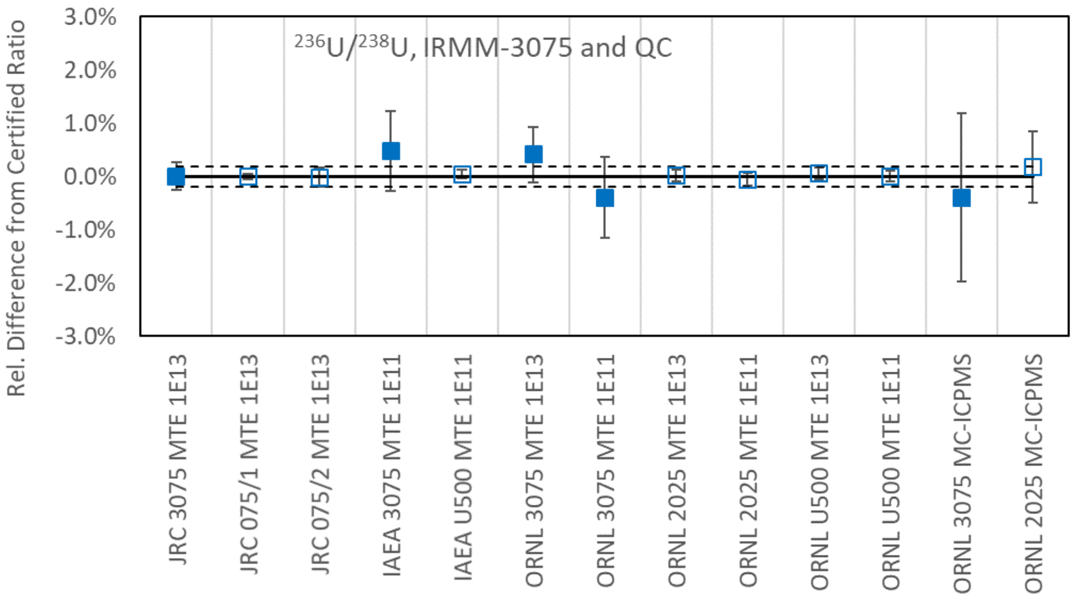
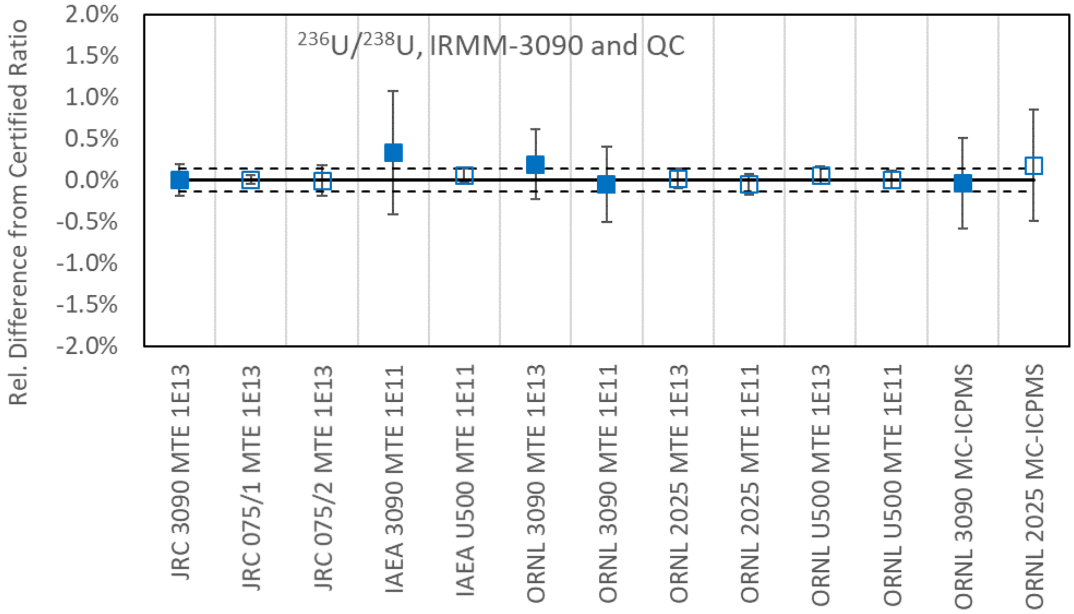


Figure 18. Verification of $n(^{236}U)/n(^{238}U)$ ratio for IRMM-3090 (■) by the IAEA/NML and ORNL, complemented by QC data (□) from the IAEA/NML, ORNL and JRC-Geel. The uncertainties are given with coverage factor k=2.



The results for the relative deviations from the certified values show that all $n(^{234}U)/n(^{238}U)$, $n(^{235}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$ ratios measured by the IAEA/NML, IAEA/ESL and ORNL demonstrate agreement with the certified values proposed by JRC-Geel within the stated uncertainties. In addition to the IRMM-3000 series, also the results for various different reference materials used as QC samples by JRC, the IAEA and ORNL agree well with the respective certified values.

In case of the minor ratios $n(^{234}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$, the uncertainties for the IRMM-3000 series are much larger compared to those obtained for the QC samples. This is due to the fact, that for the IRMM-3000 series, the minor isotope ratios are quite low but the tailing contributions from the major isotopes ^{235}U and ^{238}U towards the minor isotopes ^{234}U and ^{236}U cause significantly higher uncertainties compared to the QC samples. For the minor ratios, no results using MC-ICPMS were presented by the IAEA/ESL, because at that time the measurements of HEU samples were still under development and not yet approved as reportable data.

For the major ratios $n(^{235}U)/n(^{238}U)$, the uncertainties are on a similar level for all laboratories, and due to the internal correction, they are slightly smaller for DS measurements compared to MTE measurements. The uncertainties for the relative deviation from the certified values are a bit larger for the $n(^{235}U)/n(^{238}U)$ ratios of the NBL U500 QC samples, this is due to the larger uncertainty of 0.1% for the certified $n(^{235}U)/n(^{238}U)$ ratio.

In Figure 19 and Figure 20, QC charts are presented to show that the major ratios $n(^{235}U)/n(^{238}U)$ measured using the TIMS/DS method are stable over time periods of at least ten years. This can be used to justify the ten years validity time for certified isotope ratios obtained using this method. In particular, for the gravimetrically prepared IRMM-074 series (basic mixture of ^{235}U and ^{238}U oxides, [4]), the QC chart shows the level of accuracy (trueness) for TIMS/DS measurements using the IRMM-3636a double spike reference material, which is also used for the certification of the IRMM-3035.

In Figure 21, a QC chart for $n(^{236}U)/n(^{238}U)$ ratio measurements of the gravimetrically prepared IRMM-075/1 [5] with a certified value of $n(^{236}U)/n(^{238}U)$ = 0.000104433(37) using TIMS/MTE by JRC-Geel is shown. The precision is obviously better and the uncertainties lower when 10^{13} Ω amplifiers are used, as explained in detail in [12].

In Figure 22, a QC chart for TIMS/MTE measurements of the $n(^{234}U)/n(^{238}U)$ ratio of the IRMM-075 series [5], with a measured value of $n(^{234}U)/n(^{238}U)$ = 0.000053277(37), is shown. This ratio is the same for all six mixtures IRMM-075/1-6 and was not certified but measured using TIMS/MTE at the time of the certification for the $n(^{236}U)/n(^{238}U)$ ratios (2006). Due to the lack of highly enriched ^{234}U materials, there are no gravimetrically prepared reference materials available with certified $n(^{234}U)/n(^{238}U)$ ratios. The situation is fortunately better for the $n(^{233}U)/n(^{238}U)$ and $n(^{235}U)/n(^{238}U)$ ratios for which the gravimetrically prepared

IRMM-074 series [4] is available, and for $n(^{236}U)/n(^{238}U)$ for which the IRMM-075 series is available [5]. Since the mass of the minor isotope ^{234}U is between the minor isotopes ^{233}U and ^{236}U and the measurement techniques are similar, this might not constitute a problem, but a verification of measured $n(^{234}U)/n(^{238}U)$ ratios using a method that is independent on mass spectrometry would still be considered valuable.

As one possible resolution, the secular equilibrium value for the $n(^{234}U)/n(^{238}U)$ ratio for the ^{238}U decay chain can be calculated using the half live values for ^{234}U and ^{238}U obtained using radiometric methods, as presented in [19] for ^{234}U and in [20] for ^{238}U, to be 0.00005494(15). For certain geological formations such as calcites and well-preserved zircons a closed system can be assumed as explained by Cheng at al [11], who determined a secular equilibrium value for $n(^{234}U)/n(^{238}U)$ of 0.000054970(19) by MC-ICPMS using the IRMM-074 series for instrument calibration. The relative difference of this ratio from the ratio calculated independently using the half-lives is 0.05(27)% and therefore insignificant. This serves as a confirmation of the mass spectrometric result of Cheng at al for the $n(^{234}U)/n(^{238}U)$ ratio by an independent method, as suggested. This independent confirmation can be "extended" to $n(^{234}U)/n(^{238}U)$ ratio measurements performed by TIMS/MTE at JRC-Geel, using comparative measurements of the NBL CRM 112A uranium isotope reference material. Measurements of NBL CRM 112A using MC-ICPMS were performed by Cheng at al [11], with a $n(^{234}U)/n(^{238}U)$ ratio of 0.000052852(15), which are in excellent agreement with the $n(^{234}U)/n(^{238}U)$ ratio of 0.000052853(16) obtained using TIMS/MTE at JRC-Geel [21]. This independent confirmation for the mass spectrometric results using the radiometric ratio of the half-lives is provided at an uncertainty level of about 0.3%, which is by a factor of about 10-20 inferior to the known uncertainties from gravimetrically prepared reference materials, but this might improve as half-life values with smaller uncertainties can be provided in the future.

Figure 19. QC chart for $n(^{235}U)/n(^{238}U)$ ratio measurements of IRMM-184 using TIMS/DS by JRC-Geel. The uncertainties are given with coverage factor k=2.

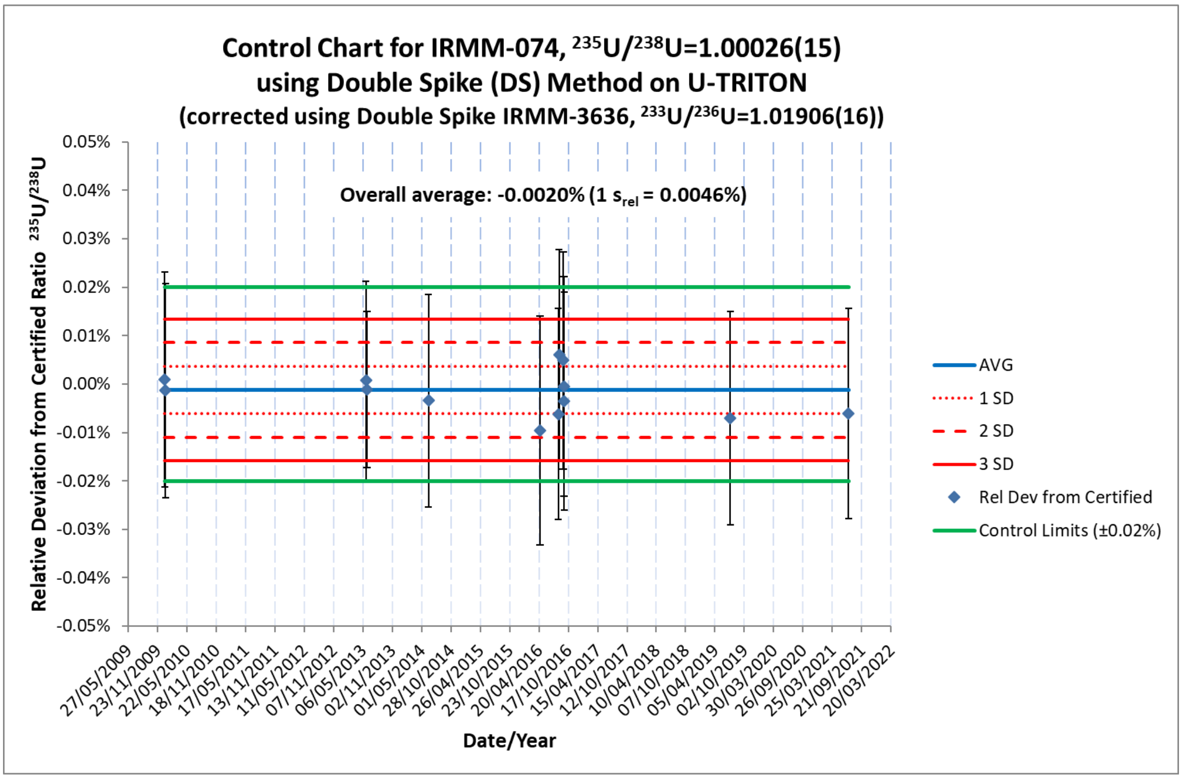


Figure 20. QC chart for $n(^{235}U)/n(^{238}U)$ ratio measurements of the gravimetrically prepared IRMM-074 ($^{235}U/^{238}U$ mixture used for preparation of the IRMM-074 series [4] using TIMS/DS by JRC-Geel. The uncertainties are given with coverage factor k=2.

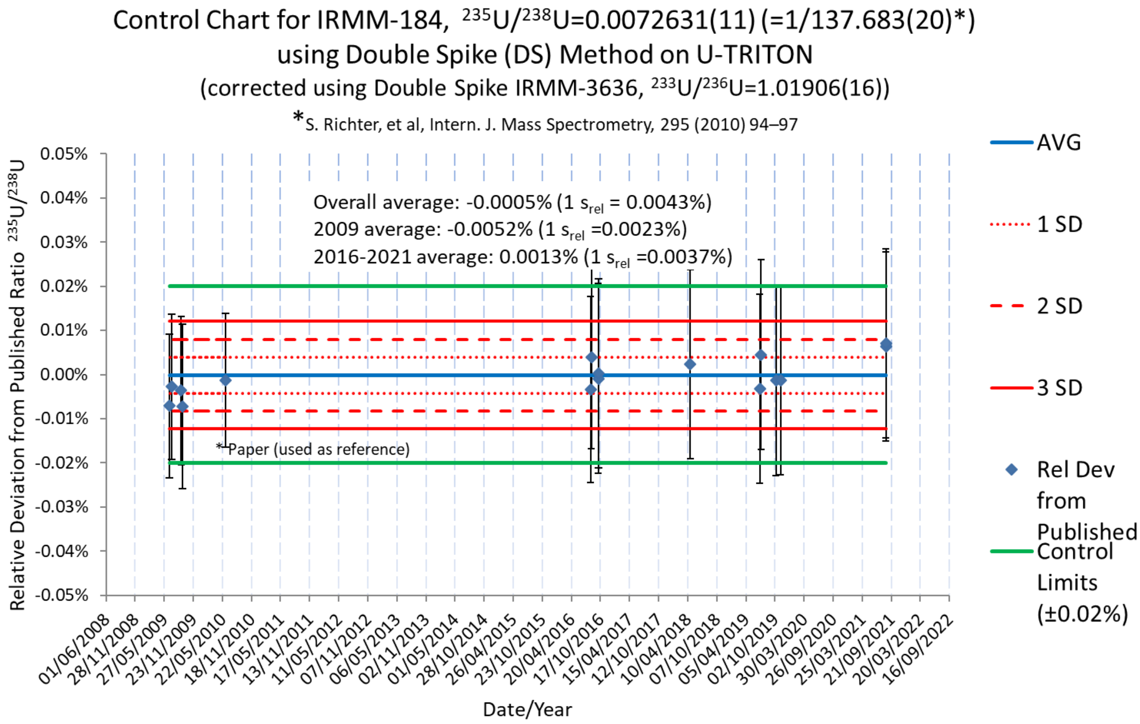


Figure 21. QC chart for $n(^{236}U)/n(^{238}U)$ ratio measurements of the gravimetrically prepared IRMM-075/1 [5], $n(^{236}U)/n(^{238}U)$ = 0.000104433(37), using TIMS/MTE by JRC-Geel. The uncertainties are given with coverage factor k=2.

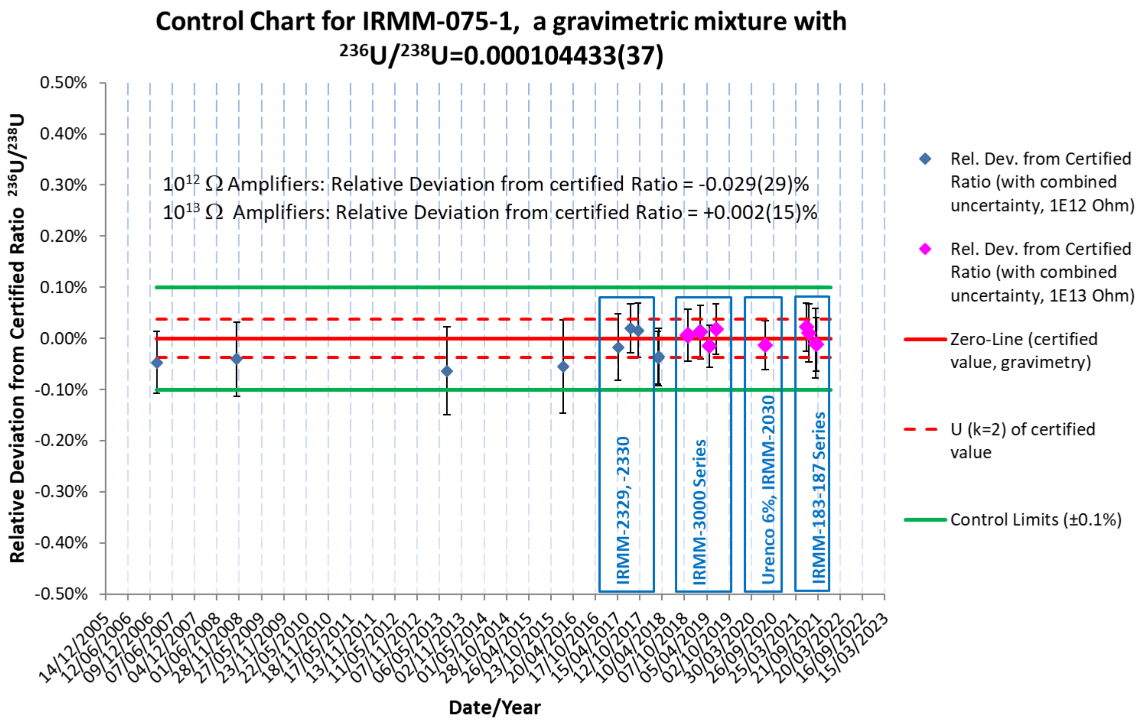
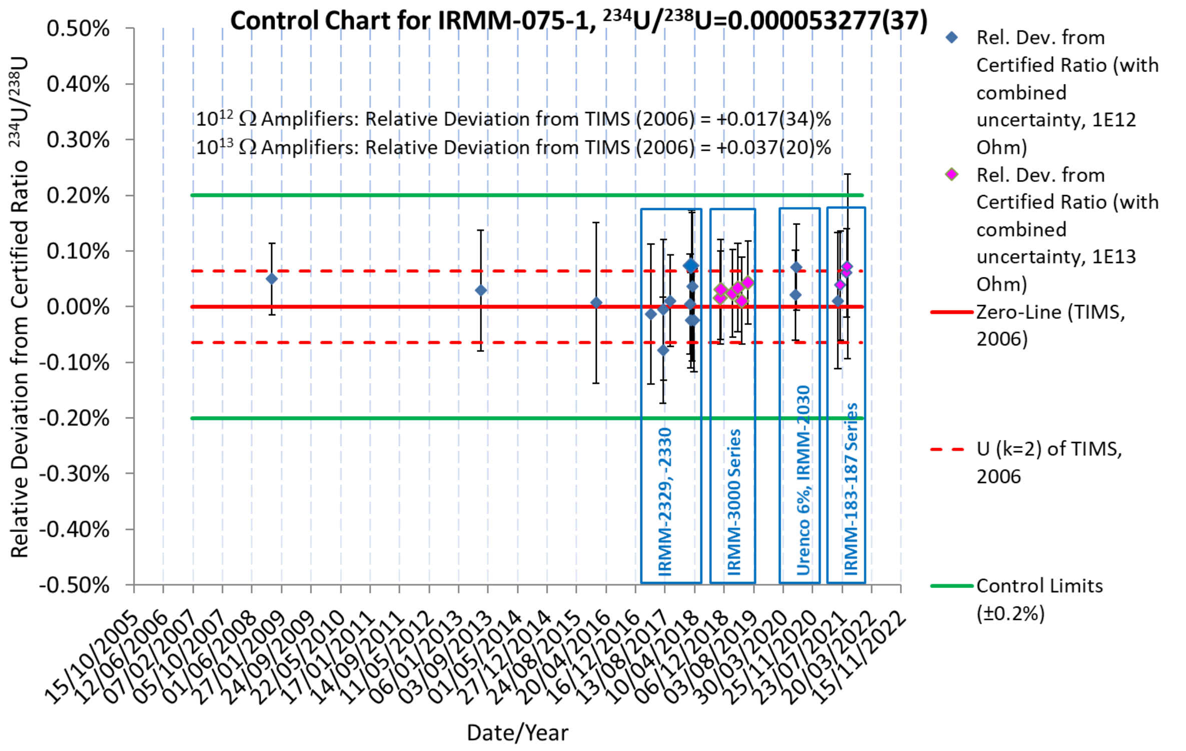


Figure 22. QC chart for $n(^{234}U)/n(^{238}U)$ ratio measurements of the IRMM-075 series [5], $n(^{234}U)/n(^{238}U)$ = 0.000053277(37), using TIMS/MTE by JRC-Geel. The uncertainties are given with coverage factor k=2.



4 Value assignment

Certified values are values that fulfil the highest standards of accuracy.

Usually the assigned uncertainties consist of uncertainties relating to characterisation, u_{char} (section 2.10), potential within-unit and between-unit inhomogeneities, combined and expressed as u_{hom} (section 2.11), potential degradation during transport, u_{trn} (section 2.12.1), and potential degradation during storage, u_{lts} (section 2.12.2). These different contributions were combined to estimate the relative expanded uncertainty of the certified value (U_{CRM}) with a coverage factor k given as:

$$\boldsymbol{U}_{\mathbf{CRM}} = \boldsymbol{k} \cdot \sqrt{\boldsymbol{u}_{\mathbf{char}}^2 + \boldsymbol{u}_{\mathbf{hom}}^2 + \boldsymbol{u}_{\mathbf{trn}}^2 + \boldsymbol{u}_{\mathbf{lts}}^2} \qquad \text{Equation 1}$$

As explained in sections 2.11 and 2.12, no assessments of the homogeneity nor of the stability were deemed necessary for the highly enriched uranium nitrate solutions, and therefore the uncertainties u_{hom} u_{trn} and u_{lts} are zero. Consequently, only the uncertainties from the characterization u_{char} have to be taken into account for the certification of the uranium isotope amount ratios in the IRMM-3000 series.

4.1 Uranium isotope amount ratios, molar masses, amount and mass fractions

The results of the characterization of the uranium isotope amount ratios for the IRMM-3000 series are presented within section 2.10 in Table 9. These results are assigned as the certified values. The uranium isotope amount ratios are given in mol·mol^{-1}.

From the certified values for the uranium isotope amount ratio, the isotope amount fractions can be derived by dividing the isotope amount ratios for the various isotopes by the sum of the isotope amount ratios:

$$\boldsymbol{n}({}^{x}\mathbf{U})/\boldsymbol{n}(\mathbf{U}) = \frac{\boldsymbol{n}({}^{x}\mathbf{U})/\boldsymbol{n}({}^{238}\mathbf{U})}{\sum_{i=\{234;235;236;238\}}^{i} \boldsymbol{n}({}^{i}\mathbf{U})/\boldsymbol{n}({}^{238}\mathbf{U})} \qquad \text{Equation 2}$$

Moreover, the molar mass of the uranium can be calculated by multiplication of the isotope amount fractions with the molar mass of the respective isotope:

$$\boldsymbol{M}(\mathbf{U}) = \sum_{i=\{234;235;236;238\}}^{i} \boldsymbol{n}({}^{i}\mathbf{U})/\boldsymbol{n}(\mathbf{U}) \cdot \boldsymbol{M}({}^{i}\mathbf{U}) \qquad \text{Equation 3}$$

For the calculation of the uranium molar mass, the molar mass of the individual isotopes of uranium (^{234}U, ^{235}U, ^{236}U and ^{238}U) have been taken from the most recent atomic mass evaluation (AME2016, [22]).

Finally, the isotope mass fractions for the different uranium isotopes can be calculated by multiplication of the isotope amount fraction with the isotope molar mass, followed by division by the molar mass of the uranium:

$$\boldsymbol{m}({}^{x}\mathbf{U})/\boldsymbol{m}(\mathbf{U}) = \frac{\boldsymbol{n}({}^{x}\mathbf{U})/\boldsymbol{n}(\mathbf{U}) \cdot \boldsymbol{M}({}^{x}\mathbf{U})}{\boldsymbol{M}(\mathbf{U})} \qquad \text{Equation 4}$$

The certified and calculated derived values related to the uranium isotopic composition for the IRMM-3000 series are summarised in Table 12 and calculations presented in Annex 4 to Annex 8.

Table 12. Certified Values for IRMM-3000 series

	$n(^{234}U)/n(^{238}U)$	**$n(^{235}U)/n(^{238}U)$**	**$n(^{236}U)/n(^{238}U)$**	
IRMM-3020	0.000004931(35)	0.254264(41)	0.000009805(63)	
IRMM-3035	0.000010669(69)	0.548531(89) (*)	0.000021224(86)	
IRMM-3050	0.00001991(13)	1.02359(18)	0.00003969(12)	
IRMM-3075	0.00005473(35)	2.81872(44)	0.00010966(20)	
IRMM-3090	0.00012770(83)	6.57731(92)	0.00025654(34)	
	$n(^{234}U)/n(U)$	**$n(^{235}U)/n(U)$**	**$n(^{236}U)/n(U)$**	**$n(^{238}U)/n(U)$**
IRMM-3020	0.000003931(28)	0.202717(26)	0.000007818(50)	0.797271(26)

IRMM-3035	0.000006890(45)	0.354220(37)	0.000013706(56)	0.645760(37)
IRMM-3050	0.000009838(63)	0.505814(44)	0.000019615(60)	0.494157(44)
IRMM-3075	0.000014332(91)	0.738100(30)	0.000028716(53)	0.261857(30)
IRMM-3090	0.00001685(11)	0.867983(16)	0.000033854(46)	0.131966(16)
	$m(^{234}U)/m(U)$	**$m(^{235}U)/m(U)$**	**$m(^{236}U)/m(U)$**	**$m(^{238}U)/m(U)$**
IRMM-3020	0.000003875(27)	0.200670(26)	0.000007772(50)	0.799318(26)
IRMM-3035	0.000006804(44)	0.351317(37)	0.000013651(56)	0.648662(37)
IRMM-3050	0.000009735(63)	0.502636(44)	0.000019574(60)	0.497335(44)
IRMM-3075	0.000014224(91)	0.735636(30)	0.000028742(53)	0.264321(30)
IRMM-3090	0.00001675(11)	0.866520(16)	0.000033941(46)	0.133429(16)
	Molar Mass			
IRMM-3020	237.441214(78)			
IRMM-3035	236.98564(11)			
IRMM-3050	236.52980(13)			
IRMM-3075	235.831309(91)			
IRMM-3090	235.440749(48)			

4.2 Diluted solutions: the IRMM-3000a series

It is planned to prepare diluted solutions of IRMM-3020, IRMM-3035, IRMM-3050, IRMM-3075 and IRMM-3090 with mass fractions in the range of 50 µg/mL - 100 µg/mL, to be named IRMM-3020a, IRMM-3035a, IRMM-3050a, IRMM-3075a and IRMM-3090a. The diluted solutions will be dispensed into screw-cap quartz ampoules and offered for sale to interested customers, who would prefer the diluted solutions for measurements by MC-ICPMS or by TIMS using the "classical" total evaporation technique.

After the dilution and dispensing, a "note to the certification file" will be prepared, containing the results of "process control measurements" (i.e. verification measurements) performed on the solutions of IRMM-3020a, IRMM-3035a, IRMM-3050a, IRMM-3075a and IRMM-3090a. If the verification measurements show agreement with the certified values for IRMM-3020, IRMM-3035, IRMM-3050, IRMM-3075 and IRMM-3090 published in this report, the same certified values will be assigned to IRMM-3020a, IRMM-3035a, IRMM-3050a, IRMM-3075a and IRMM-3090a.

5 Metrological traceability and commutability

5.1 Metrological traceability

Traceability of the obtained results is based on the traceability of all relevant input factors. Instruments were verified and calibrated with tools ensuring traceability to the International System of units (SI).

The certified values for the uranium isotope amount ratios of IRMM-3020, IRM-3050, IRMM-3075 and IRMM-3090 are traceable to the International System of Units (SI) due to the gravimetrical preparation and mixing of the ^{235}U and ^{238}U starting materials, and in case of IRMM-3035, due to the certified $n(^{233}U)/n(^{236}U)$ isotope ratio of the gravimetrically prepared double spike reference material IRMM-3636a (see traceability statement on the certificate in Annex 1).

5.2 Commutability

Many measurement procedures include one or more steps which select specific analytes from the sample for the subsequent whole measurement process. Often the complete identity of these “intermediate analytes” is not fully known or taken into account. Therefore, it is difficult to mimic all analytically relevant properties of real samples within a CRM. The degree of equivalence in the analytical behaviour of real samples and a CRM with respect to various measurement procedures is summarised in a concept called 'commutability of a reference material'. There are various definitions that define this concept. For instance, the CLSI Guideline C53-A [23] recommends the use of the following definition for the term commutability:

"The equivalence of the mathematical relationships among the results of different measurement procedures for an RM and for representative samples of the type intended to be measured."

The commutability of a CRM defines its fitness for use and is therefore a crucial characteristic when applying different measurement methods. When the commutability of a CRM is not established, the results from routinely used methods cannot be legitimately compared with the certified value to determine whether a bias does not exist in calibration, nor can the CRM be used as a calibrant.

This reference material is tailor-made to be used by the nuclear safeguards community as calibrant, QC sample and reference material mainly for mass spectrometry analysis. The commutability is being demonstrated in section 3 about verification measurements by external collaborators.

6 Instructions for use

6.1 Safety information

The IRMM-3000 series contains low-level radioactive material. The ampoules should be handled with great care and by experienced personnel in a laboratory suitably equipped for the safe handling of radioactive materials.

6.2 Storage conditions

The material should be stored. at room temperature in an upright position. However, the European Commission cannot be held responsible for changes that happen during storage of the material at the customer's premises, especially of opened samples.

6.3 Use of the certified values

The main purpose of these materials is to assess method performance for mass spectrometry, i.e. for checking accuracy of analytical results and for calibration. As any reference material, they can be used for establishing control charts or validation studies.

6.4 Use as a calibrant

The uncertainty of the certified value shall be taken into account in the estimation of the measurement uncertainty.

Comparing an analytical result with the certified value:

A result is unbiased if the combined standard uncertainty of measurement and certified value covers the difference between the certified value and the measurement result (see also *ERM Application Note 1, www.erm-crm.org*).

When assessing the method performance, the measured values of the CRMs are compared with the certified values. The procedure is summarised here:

1. Calculate the absolute difference between mean measured value and the certified value (Δ_{meas}).
2. Combine the measurement uncertainty (u_{meas}) with the uncertainty of the certified value (u_{CRM}): $u_\Delta = \sqrt{u_{\mathrm{meas}}^2 + u_{\mathrm{CRM}}^2}$.
3. Calculate the expanded uncertainty (U_Δ) from the combined uncertainty (u_Δ) using an appropriate coverage factor, corresponding to a level of confidence of approximately 95 %.
4. If $\Delta_{meas} \leq U_\Delta$ then no significant difference exists between the measurement result and the certified value, at a confidence level of approximately 95 %.

6.5 Use in quality control charts

The materials can be used for quality control charts. Using CRMs for quality control charts has the added value that a trueness assessment is built into the chart.

References

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List of abbreviations and definitions

ASTM	American Society for testing and Materials
CRM	Certified reference material
DS	Double Spike Method (for TIMS)
ESL	Environmental Sample Laboratory of the IAEA
EC	European Commission
HEU	Highly enriched Uranium
IAEA	International Atomic Energy Agency
ID	Isotope dilution
IDMS	Isotope dilution mass spectrometry
JRC	Joint Research Centre
k	Coverage factor
LEU	Low Enriched Uranium
m	Mass of substancelation
M	Molar mass
MC-ICPMS	Multi-collector inductively coupled plasma mass spectrometry
MTE	Modified total evaporation
n	Amount of substance/number or replicates
N_A	Avogadro's constant
NML	Nuclear Material Laboratory of the IAEA
R	Isotope amount ratio
RM	Reference material
SGAS	Safeguards Analytical Services of the IAEA
SI	International system of units
THF	Tetrahydrofuran
TIMS	Thermal ionisation mass spectrometry
U	Expanded uncertainty
u	Standard uncertainty
u_{char}	Standard uncertainty due to material characterisation
u_{CRM}	Combined standard uncertainty of the certified value
u_{hom}	Standard uncertainty of the homogeneity study
u_{lts}	Standard uncertainty of the long-term stability
u_{trn}	Standard uncertainty due to short-term stability (transport)
VIM	International vocabulary on metrology

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Annexes

Annex 1. Certificates of reference materials, IRMM-184, IRMM-3636a, IRMM-074, IRMM-075

EUROPEAN COMMISSION
JOINT RESEARCH CENTRE

Directorate G – Nuclear Safety and Security
G.2 – Standards for Nuclear Safety, Security and Safeguards Unit

CERTIFIED REFERENCE MATERIAL
IRMM – 184

CERTIFICATE OF ANALYSIS

Uranium in nitric acid solution		
	Isotope amount ratios	
	Certified value [1)] [mol/mol]	Uncertainty [2)] [mol/mol]
$n(^{233}U)/n(^{238}U)$	< 0.000000002	/
$n(^{234}U)/n(^{238}U)$	0.000053138	0.000000032
$n(^{235}U)/n(^{238}U)$	0.0072623	0.0000022
$n(^{236}U)/n(^{238}U)$	0.00000012446	0.00000000053

[1)] The certified values are traceable to the International System of units (SI).

[2)] The uncertainty is the expanded uncertainty with a coverage factor $k = 2$ corresponding to a level of confidence of about 95 % estimated in accordance with ISO/IEC Guide 98-3, Guide to the Expression of Uncertainty in Measurement (GUM:1995), ISO, 2008.

There is no minimum sample intake to be taken into account.

The certificate is valid for 3 years; the validity may be extended after further tests on the stability of the material are carried out.

The certificate is a revision of the original certificate of 1987, which was revised in 1993, 1999 and 2005.

Geel, September 2019

Signed: 18 SEP. 2019

Dr. Arjan Plompen
European Commission
Joint Research Centre
Directorate G – Nuclear Safety and Security
G.2 – Standards for Nuclear Safety, Security and Safeguards Unit
Retieseweg 111
B-2440 Geel, Belgium

The following values were assigned (continued):

	Isotopic mass fractions (·100)	
	Value [3) [g/g]	Uncertainty [4) [g/g]
$m(^{233}U)/m(U)$	< 0.0000002	/
$m(^{234}U)/m(U)$	0.0051868	0.0000032
$m(^{235}U)/m(U)$	0.71191	0.00021
$m(^{236}U)/m(U)$	0.000012253	0.000000052
$m(^{238}U)/m(U)$	99.28289	0.00021
	Isotopic amount fractions (·100)	
	Value [3) [mol/mol]	Uncertainty [4) [mol/mol]
$n(^{233}U)/n(U)$	< 0.0000002	/
$n(^{234}U)/n(U)$	0.0052752	0.0000032
$n(^{235}U)/n(U)$	0.72096	0.00021
$n(^{236}U)/n(U)$	0.000012356	0.000000052
$n(^{238}U)/n(U)$	99.27376	0.00022
	Molar mass	
	Value [3) [g/mol]	Uncertainty [4) [g/mol]
U	238.0288936	0.0000079

[3) The derived certified values are calculated from the certified uranium isotope amount ratios and the atomic masses according to G. Audi et al. (The 1993 atomic mass evaluation, Nuclear Physics, A565, 1-65, 1993).

[4) The uncertainty is the expanded uncertainty with a coverage factor $k = 2$ corresponding to a level of confidence of about 95 % estimated in accordance with ISO/IEC Guide 98-3, Guide to the Expression of Uncertainty in Measurement (GUM:1995), ISO, 2008.

DESCRIPTION OF THE MATERIAL

The IRMM-184 is a uranium isotopic Certified Reference Material (CRM) supplied with isotope amount ratios as certified above. IRMM-184 comes in a flame-sealed glass ampoule containing about 0.004 mol uranium in 5 mL of nitric acid solution. The concentration of nitric acid is 5 $mol \cdot L^{-1}$.

ANALYTICAL METHODS USED FOR CERTIFICATION

The certified $n(^{235}U)/n(^{238}U)$ ratio was established by mass spectrometric measurements using the MAT511 mass spectrometer. The certified $n(^{234}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$ ratios were measured using the Triton thermal ionisation mass spectrometer.
Compared to the last revision in 2005, the uncertainties for the $n(^{236}U)/n(^{238}U)$ ratios were updated according to the recommendation from ASTM C1832, section 14.6.4, by adding a relative uncertainty contribution of 0.4 % ($k = 2$) for the use of the secondary electron multiplier for measuring ^{236}U.

SAFETY INFORMATION

The IRMM-184 contains radioactive material. The ampoules should be handled with great care and by experienced personnel in a laboratory suitably equipped for the safe handling of radioactive materials.

INSTRUCTIONS FOR USE AND INTENDED USE

The material is used for calibration of mass spectrometers.

STORAGE

The vials should be stored at + 18 °C ± 5 °C.
However, the European Commission cannot be held responsible for changes that happen during storage of the material at the customer's premises, especially of opened samples.

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CERTIFIED REFERENCE MATERIAL
IRMM – 3636a

CERTIFICATE OF ANALYSIS

Uranium in nitric acid solution		
	Isotope amount content	
	Certified value 1) [µmol/g solution]	Uncertainty 2) [µmol/g solution]
^{236}U	0.211906	0.000026
	Isotope amount ratios	
	Certified value 1) [mol/mol]	Uncertainty 2) [mol/mol]
$n(^{233}U)/n(^{236}U)$	1.01906	0.00016
$n(^{234}U)/n(^{236}U)$	0.00036606	0.00000048
$n(^{235}U)/n(^{236}U)$	0.000045480	0.000000074
$n(^{238}U)/n(^{236}U)$	0.00023481	0.00000038

1) The certified values are traceable to the International System of units (SI). The reference date for the certified values is July 1, 2007.

2) The uncertainty is the expanded uncertainty with a coverage factor $k = 2$ corresponding to a level of confidence of about 95 % estimated in accordance with ISO/IEC Guide 98-3, Guide to the Expression of Uncertainty in Measurement (GUM:1995), ISO, 2008.

There is no minimum sample intake to be taken into account.

The certificate is valid for 3 years; the validity may be extended after further tests on the stability of the material are carried out.

The certificate is a revision of the original certificate of 2008, which was revised in 2009.

Geel, September 2019

1 8 SEP 2019

Signed:

Dr. Arjan Plompen
European Commission
Joint Research Centre
Directorate G – Nuclear Safety and Security
G.2 – Standards for Nuclear Safety, Security and Safeguards Unit
Retieseweg 111
B-2440 Geel, Belgium

1 8 SEP. 2019

The following values were assigned (continued):

	Isotopic mass fractions (·100)	
	Value 3) [g/g]	Uncertainty 4) [g/g]
$m(^{233}U)/m(U)$	50.1355	0.0039
$m(^{234}U)/m(U)$	0.018087	0.000024
$m(^{235}U)/m(U)$	0.0022568	0.0000037
$m(^{236}U)/m(U)$	49.8324	0.0039
$m(^{238}U)/m(U)$	0.011801	0.000019
	Isotopic amount fractions (·100)	
	Value 3) [mol/mol]	Uncertainty 4) [mol/mol]
$n(^{233}U)/n(U)$	50.4558	0.0039
$n(^{234}U)/n(U)$	0.018125	0.000024
$n(^{235}U)/n(U)$	0.0022518	0.0000037
$n(^{236}U)/n(U)$	49.5122	0.0039
$n(^{238}U)/n(U)$	0.011626	0.000019
	Amount content	
	Value 3) [µmol/g solution]	Uncertainty 4) [µmol/g solution]
U	0.427988	0.000054
^{233}U	0.215945	0.000035
	Mass fraction	
	Value 3) [mg/g solution]	Uncertainty 4) [mg/g solution]
U	0.100375	0.000013
^{233}U	0.0503237	0.0000081
^{236}U	0.0500195	0.0000062
	Molar mass	
	Value 3) [g/mol]	Uncertainty 4) [g/mol]
U	234.52874	0.00012

3) The derived certified values are calculated from the certified amount content of ^{236}U, uranium isotope amount ratios and the atomic masses according to G. Audi et al. (The 2003 atomic mass evaluation, Nuclear Physics, A729, 337-676, 2003). The reference date for the derived certified values is July 1, 2007.

4) The uncertainty is the expanded uncertainty with a coverage factor k = 2 corresponding to a level of confidence of about 95 % estimated in accordance with ISO/IEC Guide 98-3, Guide to the Expression of Uncertainty in Measurement (GUM:1995), ISO, 2008.

DESCRIPTION OF THE MATERIAL

The IRMM-3636a is a uranium spike Certified Reference Material (CRM) supplied with an isotope amount content of ^{236}U and isotope amount ratios as certified above. IRMM-3636a is a dilution of IRMM-3636 and comes in a flame-sealed quartz ampoule containing about 0.42 µmol uranium in about 1 mL of nitric acid solution. The concentration of nitric acid is 1 $mol \cdot L^{-1}$.

ANALYTICAL METHODS USED FOR CERTIFICATION

The certified values are based on the metrological dilution of IRMM-3636, which was prepared by the gravimetric mixing of highly enriched ^{233}U and ^{236}U starting solutions and verified by isotope dilution mass spectrometry (IDMS). The isotope ratio measurements were performed on a Triton Thermal Ionisation Mass Spectrometer and calibrated by means of synthetic uranium isotope mixtures.

SAFETY INFORMATION

The IRMM-3636a contains radioactive material. The ampoules should be handled with great care and by experienced personnel in a laboratory suitably equipped for the safe handling of radioactive materials.

INSTRUCTIONS FOR USE AND INTENDED USE

This spike Certified Reference Material (CRM) is used as a calibrant to determine the uranium amount content by isotope dilution mass spectrometry (IDMS).

STORAGE

The vials should be stored at + 18 °C ± 5 °C.
However, the European Commission cannot be held responsible for changes that happen during storage of the material at the customer's premises, especially of opened samples.

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NOTE

A technical report on the preparation and certification of IRMM-3636a is available on the internet (https://crm.jrc.ec.europa.eu/). A paper copy can be obtained from JRC - Geel on request.

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EUROPEAN COMMISSION
JOINT RESEARCH CENTRE

Directorate G – Nuclear Safety and Security
G.2 – Standards for Nuclear Safety, Security and Safeguards Unit

CERTIFIED REFERENCE MATERIAL
IRMM – 074

CERTIFICATE OF ANALYSIS

Uranium in nitric acid solution

Code number	Certified isotope amount ratios[1] [mol/mol]		
	$n(^{233}U)/n(^{235}U)$ U = 0.025 % (relative) [2]	$n(^{233}U)/n(^{238}U)$ U = 0.025 % (relative) [2]	$n(^{235}U)/n(^{238}U)$ U = 0.015 % (relative) [2]
IRMM-074/1	1.02685	1.02711	1.000254
IRMM-074/2	0.307993	0.308072	1.000258
IRMM-074/3	0.0102288	0.0102314	1.000259
IRMM-074/4	0.00307358	0.00307437	1.000259
IRMM-074/5	0.00103061	0.00103088	1.000259
IRMM-074/6	0.000307778	0.000307858	1.000259
IRMM-074/7	0.000102603	0.000102629	1.000259
IRMM-074/8	0.0000308011	0.0000308091	1.000259
IRMM-074/9	0.0000081587	0.0000081608	1.000259
IRMM-074/10	0.00000101886	0.00000101913	1.000259

[1] The certified values are traceable to the International System of units (SI). The reference date for the certified values is June 2005.
[2] The uncertainty (U) is the relative expanded uncertainty with a coverage factor k = 2 corresponding to a level of confidence of about 95 % estimated in accordance with ISO/IEC Guide 98-3, Guide to the Expression of Uncertainty in Measurement (GUM:1995), ISO, 2008.

There is no minimum sample intake to be taken into account.

The certificate is valid for 10 years; the validity may be extended after further tests on the stability of the material are carried out. The certificate is a revision of the original certificate of 2010.

Geel, September 2019

Signed: 18 SEP. 2019

Dr. Arjan Plompen
European Commission
Joint Research Centre
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G.2 – Standards for Nuclear Safety, Security and Safeguards Unit
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B-2440 Geel, Belgium

DESCRIPTION OF THE MATERIAL

The IRMM-074 is a uranium isotopic Certified Reference Material (CRM) supplied with isotope amount ratios as certified above. The IRMM-074 consists of a set of ten units, each containing approximately 0.2 mg uranium as uranyl nitrate in 2 mL of nitric acid solution in a sealed quartz glass ampoule. The concentration of nitric acid is 1 $mol \cdot L^{-1}$.

ANALYTICAL METHODS USED FOR CERTIFICATION

The certified values were established by gravimetric mixing of highly enriched ^{233}U, ^{235}U and ^{238}U starting materials and verified by thermal ionisation mass spectrometry (TIMS).

SAFETY INFORMATION

The IRMM-074 contains radioactive material. The ampoules should be handled with great care and by experienced personnel in a laboratory suitably equipped for the safe handling of radioactive materials.

INSTRUCTIONS FOR USE AND INTENDED USE

The material is intended for the verification and correction of non-linearities of the entire mass spectrometer measurement system.

STORAGE

The vials should be stored at + 18 °C ± 5 °C.
However, the European Commission cannot be held responsible for changes that happen during storage of the material at the customer's premises, especially of opened samples.

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NOTE

A technical report on the preparation and certification of IRMM-074 is available on the internet (https://crm.jrc.ec.europa.eu/). A paper copy can be obtained from JRC - Geel on request.

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EUROPEAN COMMISSION
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Directorate G – Nuclear Safety and Security
G.2 – Standards for Nuclear Safety, Security and Safeguards Unit

CERTIFIED REFERENCE MATERIAL
IRMM – 075

CERTIFICATE OF ANALYSIS

Uranium in nitric acid solution		
Code number	Isotope amount ratios	
	$n(^{236}U)/n(^{238}U)$	
	Certified value 1) [mol/mol]	Uncertainty 2) [mol/mol]
IRMM-075/1	$1.04433 \cdot 10^{-4}$	$3.7 \cdot 10^{-8}$
IRMM-075/2	$1.14160 \cdot 10^{-5}$	$4.0 \cdot 10^{-9}$
IRMM-075/3	$1.04093 \cdot 10^{-6}$	$3.6 \cdot 10^{-10}$
IRMM-075/4	$1.13742 \cdot 10^{-7}$	$4.0 \cdot 10^{-11}$
IRMM-074/5	$1.06519 \cdot 10^{-8}$	$7.5 \cdot 10^{-12}$
IRMM-075/6	$1.0885 \cdot 10^{-9}$	$6.3 \cdot 10^{-12}$

1) The certified values are traceable to the International System of units (SI). The reference date for the certified values is May 6, 2006.

2) The uncertainty is the expanded uncertainty with a coverage factor k = 2 corresponding to a level of confidence of about 95 % estimated in accordance with ISO/IEC Guide 98-3, Guide to the Expression of Uncertainty in Measurement (GUM:1995), ISO, 2008.

There is no minimum sample intake to be taken into account.

The certificate is valid for 10 years; the validity may be extended after further tests on the stability of the material are carried out.

The certificate is a revision of the original certificate of 2007.

Geel, September 2019

Signed: ______________ 18 SEP. 2019

Dr. Arjan Plompen
European Commission
Joint Research Centre
Directorate G – Nuclear Safety and Security
G.2 – Standards for Nuclear Safety, Security and
Safeguards Unit
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B-2440 Geel, Belgium

DESCRIPTION OF THE MATERIAL

The IRMM-075 is a uranium isotopic Certified Reference Material (CRM) supplied with isotope amount ratios as certified above. The IRMM-075 consists of a set of six units, each containing approximately 1 mg uranium as uranyl nitrate in 1 mL of nitric acid solution in a sealed quartz glass ampoule. The concentration of nitric acid is 1 $mol \cdot L^{-1}$.

ANALYTICAL METHODS USED FOR CERTIFICATION

The certified values were established by gravimetric mixing of highly enriched ^{236}U and ^{nat}U starting materials and verified by thermal ionisation mass spectrometry (TIMS).

SAFETY INFORMATION

The IRMM-075 contains radioactive material. The ampoules should be handled with great care and by experienced personnel in a laboratory suitably equipped for the safe handling of radioactive materials.

INSTRUCTIONS FOR USE AND INTENDED USE

The material is intended for the verification and correction of non-linearities of the entire mass spectrometer measurement system.

STORAGE

The vials should be stored at + 18 °C ± 5 °C.
However, the European Commission cannot be held responsible for changes that happen during storage of the material at the customer's premises, especially of opened samples.

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NOTE

A technical report on the preparation and certification of IRMM-075 is available on the internet (https://crm.jrc.ec.europa.eu/). A paper copy can be obtained from JRC - Geel on request.

European Commission – Joint Research Centre
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Annex 2. Weighing Certificate E3731 for IRMM-3000 series

EUROPEAN COMMISSION Joint Research Centre	**Certificate of weighing**	irmm Institute for Reference Materials and Measurements

E. 3731 **Issued date:** 07 December 2009 **Page 1 of 1**

Applicant: Verbruggen **Group:** RM-Nuclear

Project: U- mixtures **RM-unit ref.:** New Series 3000

Description: Preparation of series 3000 mixtures U-235 and U-238.

Date of receipt of request: 14 September 2009 **Weighing date:** 23 September 2009

The reported results applies only to the objects / samples described in this certificate

ID number	g U-235 oxide	g U-238 oxide	g solution
IRMM-3020	0.26112 (3)	1.03822 (5)	253.388 (5)
IRMM-3035	0.25812 (3)	0.47673 (5)	128.574 (5)
IRMM-3050	0.29850 (3)	0.29481 (3)	103.176 (5)
IRMM-3075	0.81046 (5)	0.29067 (3)	183.006 (5)
IRMM-3090	2.25012 (10)	0.34584 (3)	423.584 (5)

Observations:

The measurements and uncertainty estimates, were performed according to working instruction WI-0185, "Mass determination by substitution weighing"on balances AT 201 and AX 504 with IRMM inventory No 1996 00547 73 and 2001 00297 70.

Traceability:

The certified mass values are traceable to the International Kilogram Prototype via regular calibrations of the IRMM principal kilogram. The set of working mass standards M 10 was used as reference in the mass determination.

Uncertainty:

All reported uncertainties are expanded uncertainties $U = k \cdot u_c$ where u_c is the combined standard uncertainty calculated according to the ISO/BIPM Guide to the expression of Uncertainty in Measurement. The coverage factor $k = 2$ corresponds to a coverage probability of about 95%. U applies to the last digit of the value of the measurement result and is given in parentheses ().

Annexes:

Signature
Mass Metrology Service

Retieseweg, B-2440 Geel, Belgium; Tel.: +32-(0)14-571 211 •Fax: +32-(0)14-571 978• http://www.irmm.jrc.be

The mission of IRMM is to promote a common and reliable European measurement system in support of EU policies.

Annex 3. Gravimetrical calculation of isotope ratios for IRMM-3020 as an example.

IRMM-3020

Two purified and enriched uranium isotopic materials - U-235 and U-238 are treated together in an oven under a controlled atmosphere (low humidity) and dissolved in nitric acid to make a mixture in solution of U-235 (BC02154) and U -238 (BC02677), 1 mg/g.

Input file for masses of U-oxides, 235U and 238U: U:\Nuclear Safeguards\Nuclear\IRMM-3000 Series - Certification Folder \Characterisation assessment \Gravimetrical Calculations\Gravimetric Calc of Ratios\E 3731 new IRMM-3000.doc

Stochiometry uncertainty set at 0.01% with correlation = 1. Impurity level set at 42 ± 23 ppm for 235U and 199 ± 98 ppm for U238, correlation = 0 Impurity data for 235U: U:\Nuclear Safeguards\Nuclear Certification Folder \Characterisation assessment \Gravimetrical Calculations \Impurities 235U base material BC02154\impurity assesment SCK IRMM.xls Impurity data for 238U: U: \Nuclear Safeguards \Nuclear\IRMM-3000 Series - Certification Folder\Characterisation assessment \Gravimetrical Calculations \Impurities 238U base material BC02677\SN3S180004 Addendum 1 - S new.xls

Input file for isotopic compositon of 235U material: U:\Nuclear Safeguards\Nuclear\IRMM-3000 Series - Certification Folder \Characterisation assessment \Gravimetrical Calculations\Gravimetric Calc of Ratios\LOT 02154 INM-10154 235-Enrich-GUMWB-2

Input file for isotopic compositon of 238U material: U:\Nuclear Safeguards\Nuclear\IRMM-3000 Series - Certification Folder \Characterisation assessment \Gravimetrical Calculations\Gravimetric Calc of Ratios\LOT 02677 238U-Enriched-GUMWB.smu

18 January 2008 by André Verbruggen

Revised: 7 January 2020, 8 May, by Stephan Richter

Model Equation:

{ Calculate stoichiometry assuming U_3O_8 and allowing an uncertainty on the oxygen content }

{set all f_O=8.0 +/- 0.01%, correlation coefficiant=1.0 }

$f_{235oxide} = 3*M_{235U}/(3*M_{SM235} + f_{O5} *M_O);$

$f_{238oxide} = 3*M_{238U}/(3*M_{SM238} + f_{O8} *M_O);$

{ Calculate mass of U from weight of oxide corrected for impurities and stoichiometry}

$m_{U235} = m_{U235O} * f_{235oxide} * (1- \delta_{U235O});$

$m_{U238} = m_{U238O} * f_{238oxide} * (1- \delta_{U238O});$

{Mixture BC02154 U235 with BC02677 U238 (E.3731)}

$n_{UfromSM235}=m_{U235}/M_{SM235};$

$n_{233\text{-}from\text{-}SM235}=f_{233\text{-}from\text{-}SM235}*n_{UfromSM235};$

$n_{234\text{-}from\text{-}SM235}=f_{234\text{-}from\text{-}SM235}*n_{UfromSM235};$

$n_{235\text{-}from\text{-}SM235}=f_{235\text{-}from\text{-}SM235}*n_{UfromSM235};$

$n_{236\text{-}from\text{-}SM235}=f_{236\text{-}from\text{-}SM235}*n_{UfromSM235};$

$n_{238\text{-}from\text{-}SM235}=f_{238\text{-}from\text{-}SM235}*n_{UfromSM235};$

$n_{UfromSM238}=m_{U238}/M_{SM238};$

$n_{233\text{-}from\text{-}SM238}=f_{233\text{-}from\text{-}SM238}*n_{UfromSM238};$

$n_{234\text{-}from\text{-}SM238}=f_{234\text{-}from\text{-}SM238}*n_{UfromSM238};$

$n_{235\text{-}from\text{-}SM238}=f_{235\text{-}from\text{-}SM238}*n_{UfromSM238};$

$n_{236\text{-}from\text{-}SM238}=f_{236\text{-}from\text{-}SM238}*n_{UfromSM238};$

$n_{238\text{-}from\text{-}SM238}=f_{238\text{-}from\text{-}SM238}*n_{UfromSM238};$

$n_{233}=n_{233\text{-from-SM235}}+n_{233\text{-from-SM238}};$
$n_{234}=n_{234\text{-from-SM235}}+n_{234\text{-from-SM238}};$
$n_{235}=n_{235\text{-from-SM235}}+n_{235\text{-from-SM238}};$
$n_{236}=n_{236\text{-from-SM235}}+n_{236\text{-from-SM238}};$
$n_{238}=n_{238\text{-from-SM235}}+n_{238\text{-from-SM238}};$

$n_{total}=n_{233}+n_{234}+n_{235}+n_{236}+n_{238};$

$R_{238/235U}=n_{238}/n_{235};$
$R_{236/235U}=n_{236}/n_{235};$
$R_{234/235U}=n_{234}/n_{235};$
$R_{233/235U}=n_{233}/n_{235};$

$R_{236/238U}=n_{236}/n_{238};$
$R_{235/238U}=n_{235}/n_{238};$
$R_{234/238U}=n_{234}/n_{238};$
$R_{233/238U}=n_{233}/n_{238};$

{---molar mass of uranium in the sample------------------}

$M_U = M_{233U}\cdot f_{233U} + M_{234U}\cdot f_{234U} + M_{235U}\cdot f_{235U} + M_{236U}\cdot f_{236U} + M_{238U}\cdot f_{238U};$

{---amount abundances in the sample----------------------}

$f_{233U} = R_{233/235U}/\Sigma R_U;$
$f_{234U} = R_{234/235U}/\Sigma R_U;$
$f_{235U} = 1/\Sigma R_U;$
$f_{236U} = R_{236/235U}/\Sigma R_U;$
$f_{238U} = R_{238/235U}/\Sigma R_U;$

$\Sigma R_U = 1 + R_{233/235U} + R_{234/235U} + R_{236/235U} + R_{238/235U};$

{--mass abundances in the sample---------------------------}

$w_{233U} = f_{233U}\cdot M_{233U}/M_U;$
$w_{234U} = f_{234U}\cdot M_{234U}/M_U;$
$w_{235U} = f_{235U}\cdot M_{235U}/M_U;$
$w_{236U} = f_{236U}\cdot M_{236U}/M_U;$
$w_{238U} = f_{238U}\cdot M_{238U}/M_U;$

List of Quantities:

Quantity	Unit	Definition
m_{U235O}	g	mass in U-235 oxide starting material BC02154 (E3731)
m_{U238O}	g	mass in U-238 oxide starting material BC02677 (E3731)
m_{U235}	g	mass in U in U-235 starting material BC02154 (E3731)
m_{U238}	g	mass in U in U-238 starting material BC02677 (E3731)
$f_{235oxide}$		stoichiometry mass fraction of U in U-235 oxide

Quantity	Unit	Definition
$f_{238oxide}$		stoichiometry mass fraction of U in U-238 oxide
f_{O5}		Oxygen content in U-235 oxide starting material BC02154
f_{O8}		Oxygen content in U-238 oxide starting material BC02155
M_O	g/mol	Atomic weight oxygen
δ_{U235O}		impurity level in U of U-235 BC02154
δ_{U238O}		impurity level in U of U-238 BC02677
$n_{UfromSM235}$	mol	amount of U from U-235 starting material
$n_{UfromSM238}$	mol	amount of U from U-238 starting material
$f_{233\text{-}from\text{-}SM235}$		amount fraction of U233 from U-235 starting material
$f_{234\text{-}from\text{-}SM235}$		amount fraction of U234 from U-235 starting material
$f_{235\text{-}from\text{-}SM235}$		amount fraction of U235 from U-235 starting material
$f_{236\text{-}from\text{-}SM235}$		amount fraction of U236 from U-235 starting material
$f_{238\text{-}from\text{-}SM235}$		amount fraction of U238 from U-235 starting material
$f_{233\text{-}from\text{-}SM238}$		amount fraction of U233 from U-238 starting material
$f_{234\text{-}from\text{-}SM238}$		amount fraction of U234 from U-238 starting material
$f_{235\text{-}from\text{-}SM238}$		amount fraction of U235 from U-238 starting material
$f_{236\text{-}from\text{-}SM238}$		amount fraction of U236 from U-238 starting material
$f_{238\text{-}from\text{-}SM238}$		amount fraction of U238 from U-238 starting material
$n_{233\text{-}from\text{-}SM235}$	mol	moles of U233 from U-235 starting material
$n_{234\text{-}from\text{-}SM235}$	mol	moles of U234 from U-235 starting material
$n_{235\text{-}from\text{-}SM235}$	mol	moles of U235 from U-235 starting material
$n_{236\text{-}from\text{-}SM235}$	mol	moles of U236 from U-235 starting material
$n_{238\text{-}from\text{-}SM235}$	mol	moles of U238 from U-235 starting material
$n_{233\text{-}from\text{-}SM238}$	mol	moles of U233 from U-238 starting material
$n_{234\text{-}from\text{-}SM238}$	mol	moles of U234 from U-238 starting material
$n_{235\text{-}from\text{-}SM238}$	mol	moles of U235 from U-238 starting material
$n_{236\text{-}from\text{-}SM238}$	mol	moles of U236 from U-238 starting material
$n_{238\text{-}from\text{-}SM238}$	mol	moles of U238 from U-238 starting material
n_{233}	mol	moles of U233
n_{234}	mol	moles of U234
n_{235}	mol	moles of U235
n_{236}	mol	moles of U236
n_{238}	mol	moles of U238
n_{total}	mol	moles of U
M_U	g/mol	molar mass
M_{233U}	g/mol	atomic mass 233U
M_{234U}	g/mol	atomic mass 234U
M_{235U}	g/mol	atomic mass 235U

Quantity	Unit	Definition
M_{236U}	g/mol	atomic mass 236U
M_{238U}	g/mol	atomic mass 238U
$R_{233/238U}$	mol/mol	isotope ratio 233/238U
$R_{234/238U}$	mol/mol	isotope ratio 234/238U
$R_{235/238U}$	mol/mol	isotope ratio 235/238U
$R_{236/238U}$	mol/mol	isotope ratio 236/238U
$R_{233/235U}$	mol/mol	isotope ratio 233/235U
$R_{234/235U}$	mol/mol	isotope ratio 234/235U
$R_{236/235U}$	mol/mol	isotope ratio 236/235U
$R_{238/235U}$	mol/mol	isotope ratio 238/235U
f_{233U}		isotope amount fraction 233U
f_{234U}		isotope amount fraction 234U
f_{235U}		isotope amount fraction 235U
f_{236U}		isotope amount fraction 236U
f_{238U}		isotope amount fraction 238U
w_{233U}		isotope mass fraction 233U
w_{234U}		isotope mass fraction 234U
w_{235U}		isotope mass fraction 235U
w_{236U}		isotope mass fraction 236U
w_{238U}		isotope mass fraction 238U
ΣR_U		
M_{SM235}	g/mol	Molar Mass U235 SM material BC02154
M_{SM238}	g/mol	Molar Mass U238 SM material BC02677

m_{U235O}:
Type B normal distribution
Value: 0.26112 g
Expanded Uncertainty: 0.00003 g
Coverage Factor: 2

m_{U238O}:
Type B normal distribution
Value: 1.03822 g
Expanded Uncertainty: 0.00005 g
Coverage Factor: 2

f_{O5}:
Type B normal distribution
Value: 8
Expanded Uncertainty: 0.01 %
Coverage Factor: 2

f_{O8}:
Type B normal distribution
Value: 8
Expanded Uncertainty: 0.01 %
Coverage Factor: 2

M_O: Type B normal distribution
Value: 15.9994 g/mol
Expanded Uncertainty: 0.0003 g/mol
Coverage Factor: 2

δ_{U235O}: Type B normal distribution
Value: 0.000042
Expanded Uncertainty: 0.000023
Coverage Factor: 2

δ_{U238O}: Type B normal distribution
Value: 0.000199
Expanded Uncertainty: 0.000098
Coverage Factor: 2

$f_{233\text{-from-SM235}}$: Import
Filename: LOT 02154 INM-10154 235-Enrich-GUMWB-2.SMU
Symbol: f_{233U}

$f_{234\text{-from-SM235}}$: Import
Filename: LOT 02154 INM-10154 235-Enrich-GUMWB-2.SMU
Symbol: f_{234U}

$f_{235\text{-from-SM235}}$: Import
Filename: LOT 02154 INM-10154 235-Enrich-GUMWB-2.SMU
Symbol: f_{235U}

$f_{236\text{-from-SM235}}$: Import
Filename: LOT 02154 INM-10154 235-Enrich-GUMWB-2.SMU
Symbol: f_{236U}

$f_{238\text{-from-SM235}}$: Import
Filename: LOT 02154 INM-10154 235-Enrich-GUMWB-2.SMU
Symbol: f_{238U}

$f_{233\text{-from-SM238}}$: Import
Filename: LOT 02677 238U-Enriched-GUMWB.smu
Symbol: f_{233U}

$f_{234\text{-from-SM238}}$: Import
Filename: LOT 02677 238U-Enriched-GUMWB.smu
Symbol: f_{234U}

$f_{235\text{-from-SM238}}$: Import
Filename: LOT 02677 238U-Enriched-GUMWB.smu
Symbol: f_{235U}

$f_{236\text{-from-SM238}}$: Import
Filename: LOT 02677 238U-Enriched-GUMWB.smu
Symbol: f_{236U}

$f_{238\text{-from-SM238}}$: Import
Filename: LOT 02677 238U-Enriched-GUMWB.smu
Symbol: f_{238U}

M_{233U}: Type B normal distribution
Value: 233.0396344 g/mol
Expanded Uncertainty: 0.0000048 g/mol
Coverage Factor: 2

Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003

M_{234U}: Type B normal distribution
Value: 234.0409504 g/mol
Expanded Uncertainty: 0.0000024 g/mol
Coverage Factor: 2

Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003

M_{235U}: Type B normal distribution
Value: 235.0439282 g/mol
Expanded Uncertainty: 0.0000024 g/mol
Coverage Factor: 2

Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003

M_{236U}: Type B normal distribution
Value: 236.0455662 g/mol
Expanded Uncertainty: 0.0000024 g/mol
Coverage Factor: 1

Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003

M_{238U}: Type B normal distribution
Value: 238.0507870 g/mol
Expanded Uncertainty: 0.0000032 g/mol
Coverage Factor: 2

Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003

M_{SM235}: Import
Filename: LOT 02154 INM-10154 235-Enrich-GUMWB-2.SMU
Symbol: M_U

M_{SM238}: Import
Filename: LOT 02677 238U-Enriched-GUMWB.smu
Symbol: M_U

Input Correlation:

	$f_{234\text{-from-SM235}}$	$f_{235\text{-from-SM235}}$	$f_{236\text{-from-SM235}}$	$f_{238\text{-from-SM235}}$	M_{SM235}
$f_{234\text{-from-SM235}}$	1	-0.4125	-0.0014	-0.0069	-0.0364
$f_{235\text{-from-SM235}}$	-0.4125	1	-0.9103	-0.0157	-0.0577
$f_{236\text{-from-SM235}}$	-0.0014	-0.9103	1	0.0061	0.0798
$f_{238\text{-from-SM235}}$	-0.0069	-0.0157	0.0061	1	0.0041
M_{SM235}	-0.0364	-0.0577	0.0798	0.0041	1

	$f_{234\text{-from-SM238}}$	$f_{235\text{-from-SM238}}$	$f_{236\text{-from-SM238}}$	$f_{238\text{-from-SM238}}$	M_{SM238}
$f_{234\text{-from-SM238}}$	1	0.0529	0.0212	-0.0651	-0.0010
$f_{235\text{-from-SM238}}$	0.0529	1	0.3335	-0.9995	-0.0138
$f_{236\text{-from-SM238}}$	0.0212	0.3335	1	-0.3593	-0.0048
$f_{238\text{-from-SM238}}$	-0.0651	-0.9995	-0.3593	1	0.0138
M_{SM238}	-0.0010	-0.0138	-0.0048	0.0138	1

	f_{O5}	f_{O8}
f_{O5}	1	1
f_{O8}	1	1

Interim Results:

Quantity	**Value**	**Standard Uncertainty**
m_{U235}	0.2209942 g	$13.1\cdot10^{-6}$ g
m_{U238}	0.8802493 g	$48.5\cdot10^{-6}$ g
$f_{235oxide}$	0.84636764	$6.61\cdot10^{-6}$
$f_{238oxide}$	0.84801341	$6.56\cdot10^{-6}$
$n_{UfromSM235}$	$940.2252\cdot10^{-6}$ mol	$55.6\cdot10^{-9}$ mol
$n_{UfromSM238}$	$3.697738\cdot10^{-3}$ mol	$204\cdot10^{-9}$ mol
$n_{234\text{-from-SM235}}$	$20.0096\cdot10^{-9}$ mol	$41.0\cdot10^{-12}$ mol
$n_{235\text{-from-SM235}}$	$940.1644\cdot10^{-6}$ mol	$55.6\cdot10^{-9}$ mol
$n_{236\text{-from-SM235}}$	$39.7544\cdot10^{-9}$ mol	$90.2\cdot10^{-12}$ mol
$n_{238\text{-from-SM235}}$	$962.65\cdot10^{-12}$ mol	$1.28\cdot10^{-12}$ mol
$n_{234\text{-from-SM238}}$	$22.258\cdot10^{-12}$ mol	$332\cdot10^{-15}$ mol
$n_{235\text{-from-SM238}}$	$28.7174\cdot10^{-9}$ mol	$27.0\cdot10^{-12}$ mol
$n_{236\text{-from-SM238}}$	$345.410\cdot10^{-12}$ mol	$793\cdot10^{-15}$ mol
$n_{238\text{-from-SM238}}$	$3.697709\cdot10^{-3}$ mol	$204\cdot10^{-9}$ mol
n_{234}	$20.0318\cdot10^{-9}$ mol	$41.0\cdot10^{-12}$ mol
n_{235}	$940.1931\cdot10^{-6}$ mol	$55.6\cdot10^{-9}$ mol
n_{236}	$40.0998\cdot10^{-9}$ mol	$90.2\cdot10^{-12}$ mol
n_{238}	$3.697710\cdot10^{-3}$ mol	$204\cdot10^{-9}$ mol
n_{total}	$4.637963\cdot10^{-3}$ mol	$212\cdot10^{-9}$ mol
ΣR_U	4.932989	$315\cdot10^{-6}$

Uncertainty Budgets:

$R_{235/238U}$: **isotope ratio 235/238U**

Quantity	Value	Standard Uncertainty	Distribution	Sensitivity Coefficient	Uncertainty Contribution	Index
m_{U235O}	0.2611200 g	$15.0\cdot10^{-6}$ g	normal	0.97	$15\cdot10^{-6}$ mol/mol	51.4 %
m_{U238O}	1.0382200 g	$25.0\cdot10^{-6}$ g	normal	-0.24	$-6.1\cdot10^{-6}$ mol/mol	9.0 %
f_{O5}	8.000000	$400\cdot10^{-6}$	normal	$-4.9\cdot10^{-3}$	$-2.0\cdot10^{-6}$ mol/mol	0.0 %
f_{O8}	8.000000	$400\cdot10^{-6}$	normal	$4.8\cdot10^{-3}$	$1.9\cdot10^{-6}$ mol/mol	0.0 %
M_O	15.999400 g/mol	$150\cdot10^{-6}$ g/mol	normal	$-26\cdot10^{-6}$	$-3.9\cdot10^{-9}$ mol/mol	0.0 %
δ_{U235O}	$42.0\cdot10^{-6}$	$11.5\cdot10^{-6}$	normal	-0.25	$-2.9\cdot10^{-6}$ mol/mol	2.1 %
δ_{U238O}	$199.0\cdot10^{-6}$	$49.0\cdot10^{-6}$	normal	0.25	$12\cdot10^{-6}$ mol/mol	37.4 %
$f_{235\text{-from-}SM235}$	0.999935413	$105\cdot10^{-9}$		0.25	$27\cdot10^{-9}$ mol/mol	0.0 %
$f_{238\text{-from-}SM235}$	$1.02385\cdot10^{-6}$	$1.37\cdot10^{-9}$		-0.065	$-88\cdot10^{-12}$ mol/mol	0.0 %
$f_{235\text{-from-}SM238}$	$7.76621\cdot10^{-6}$	$7.28\cdot10^{-9}$		1.0	$7.3\cdot10^{-9}$ mol/mol	0.0 %
$f_{238\text{-from-}SM238}$	0.99999213436	$7.36\cdot10^{-9}$		-0.25	$-1.9\cdot10^{-9}$ mol/mol	0.0 %
M_{235U}	235.04392820 g/mol	$1.20\cdot10^{-6}$ g/mol	normal	$1.1\cdot10^{-3}$	$1.3\cdot10^{-9}$ mol/mol	0.0 %
M_{238U}	238.05078700 g/mol	$1.60\cdot10^{-6}$ g/mol	normal	$-1.1\cdot10^{-3}$	$-1.7\cdot10^{-9}$ mol/mol	0.0 %
M_{SM235}	235.04395228 g/mol	$1.20\cdot10^{-6}$ g/mol		$-2.0\cdot10^{-3}$	$-2.4\cdot10^{-9}$ mol/mol	0.0 %
M_{SM238}	238.05076344 g/mol	$1.60\cdot10^{-6}$ g/mol		$2.0\cdot10^{-3}$	$3.2\cdot10^{-9}$ mol/mol	0.0 %
$R_{235/238U}$	0.2542637 mol/mol	$20.4\cdot10^{-6}$ mol/mol				

Results:

Quantity	Value	Expanded Uncertainty	Coverage factor	Coverage
M_U	237.441211 g/mol	$78 \cdot 10^{-6}$ g/mol	2.00	manual
$R_{233/238U}$	0.0 mol/mol	0.0 mol/mol	2.00	manual
$R_{234/238U}$	$5.417 \cdot 10^{-6}$ mol/mol	$22 \cdot 10^{-9}$ mol/mol	2.00	manual
$R_{235/238U}$	0.254264 mol/mol	$41 \cdot 10^{-6}$ mol/mol	2.00	manual
$R_{236/238U}$	$10.844 \cdot 10^{-6}$ mol/mol	$49 \cdot 10^{-9}$ mol/mol	2.00	manual
$R_{233/235U}$	0.0 mol/mol	0.0 mol/mol	2.00	manual
$R_{234/235U}$	$21.306 \cdot 10^{-6}$ mol/mol	$87 \cdot 10^{-9}$ mol/mol	2.00	manual
$R_{236/235U}$	$42.65 \cdot 10^{-6}$ mol/mol	$190 \cdot 10^{-9}$ mol/mol	2.00	manual
$R_{238/235U}$	3.93293 mol/mol	$630 \cdot 10^{-6}$ mol/mol	2.00	manual
f_{233U}	0.0	0.0	2.00	manual
f_{234U}	$4.319 \cdot 10^{-6}$	$18 \cdot 10^{-9}$	2.00	manual
f_{235U}	0.202717	$26 \cdot 10^{-6}$	2.00	manual
f_{236U}	$8.646 \cdot 10^{-6}$	$39 \cdot 10^{-9}$	2.00	manual
f_{238U}	0.797270	$26 \cdot 10^{-6}$	2.00	manual
w_{233U}	0.0	0.0	2.00	manual
w_{234U}	$4.257 \cdot 10^{-6}$	$17 \cdot 10^{-9}$	2.00	manual
w_{235U}	0.200670	$26 \cdot 10^{-6}$	2.00	manual
w_{236U}	$8.595 \cdot 10^{-6}$	$39 \cdot 10^{-9}$	2.00	manual
w_{238U}	0.799317	$26 \cdot 10^{-6}$	2.00	manual

Annex 4. Characterization of the minor isotope ratios for IRMM-3020

	IRMM-3020	

IRMM-3020

Original folder: G:\JRC.G.2\Nuclear Safeguards\Nuclear\TRITON DATA - SHARED\IRMM-3000 Series Characterization

For 235U/238U ratios: U:\Nuclear Safeguards\Nuclear\IRMM-3000 Series - Certification Folder\Characterisation assessment \Gravimetrical Calculations \Gravimetric Calc of Ratios\IRMM-3020 Mixing NEW IMP after KA.smu

For 234U/238U and 236U/238U ratios: U:\Nuclear Safeguards\Nuclear\IRMM-3000 Series - Certification Folder \Characterisation assessment\COPY of IRMM-3000 Series Characterization\T190618 MTE - IRMM-3020 - Turret- 1B - normalized to GRAV.xls

Model Equation:

$$f_{233U} = R_{233/238U}/\Sigma R_U;$$
$$f_{234U} = R_{234/238U}/\Sigma R_U;$$
$$f_{235U} = R_{235/238U}/\Sigma R_U;$$
$$f_{236U} = R_{236/238U}/\Sigma R_U;$$
$$f_{238U} = 1/\Sigma R_U;$$
$$\Sigma R_U = R_{233/238U} + R_{234/238U} + R_{235/238U} + R_{236/238U} + 1;$$

$$M_U = M_{233U}\cdot f_{233U} + M_{234U}\cdot f_{234U} + M_{235U}\cdot f_{235U} + M_{236U}\cdot f_{236U} + M_{238U}\cdot f_{238U};$$
$$w_{233U} = f_{233U}\cdot M_{233U}/M_U;$$
$$w_{234U} = f_{234U}\cdot M_{234U}/M_U;$$
$$w_{235U} = f_{235U}\cdot M_{235U}/M_U;$$
$$w_{236U} = f_{236U}\cdot M_{236U}/M_U;$$
$$w_{238U} = f_{238U}\cdot M_{238U}/M_U;$$

List of Quantities:

Quantity	Unit	Definition
$R_{233/238U}$	mol/mol	isotope amount ratio n_{233}/n_{238} of U
$R_{234/238U}$	mol/mol	isotope amount ratio n_{234}/n_{238} of U
$R_{235/238U}$	mol/mol	isotope amount ratio n_{235}/n_{238} of U
$R_{236/238U}$	mol/mol	isotope amount ratio n_{236}/n_{238} of U
M_U		molar mass of U
f_{233U}		isotope amount fraction of ^{233}U in U
f_{234U}		isotope amount fraction of ^{234}U in U
f_{235U}		isotope amount fraction of ^{235}U in U
f_{236U}		isotope amount fraction of ^{236}U in U
f_{238U}		isotope amount fraction of ^{238}U in U
w_{233U}		isotope mass fraction of ^{233}U in U
w_{234U}		isotope mass fraction of ^{234}U in U
w_{235U}		isotope mass fraction of ^{235}U in U
w_{236U}		isotope mass fraction of ^{236}U in U
w_{238U}		isotope mass fraction of ^{238}U in U

Date: 04/07/2021	File: IRMM-3020 using major ratio from GRAV (KA).SMU	Page 1 of 3

Generated with GUM Workbench Pro Version 2.4.1.458

Quantity	Unit	Definition
M_{233U}		atomic mass for ^{233}U
M_{234U}		atomic mass for ^{234}U
M_{235U}		atomic mass for ^{235}U
M_{236U}		atomic mass for ^{236}U
M_{238U}		atomic mass for ^{238}U
ΣR_U		sum of isotope ratios for U

$R_{233/238U}$: Type B rectangular distribution
Value: 0 mol/mol
Halfwidth of Limits: 0 mol/mol

$R_{234/238U}$: Import from Excel
Filename: T190618 MTE - IRMM-3020 - Turret-1B - normalized to GRAV.xls
Worksheet: Summary-calc
Value Cell: C192 = $4.9311 \cdot 10^{-6}$ mol/mol
Standarduncertainty Cell: C193 = $17.3 \cdot 10^{-9}$ mol/mol
Degrees of Freedom Cell: G70 = 5

$R_{235/238U}$: Import
Filename: ..\Gravimetrical Calculations\Gravimetric Calc of Ratios\IRMM-3020 Mixing NEW IMP after KA.smu
Symbol: $R_{235/238U}$

$R_{236/238U}$: Import from Excel
Filename: T190618 MTE - IRMM-3020 - Turret-1B - normalized to GRAV.xls
Worksheet: Summary-calc
Value Cell: D192 = $9.8054 \cdot 10^{-6}$ mol/mol
Standarduncertainty Cell: D193 = $31.5 \cdot 10^{-9}$ mol/mol
Degrees of Freedom Cell: G70 = 5

M_{233U}: Type B normal distribution
Value: 233.0396344
Expanded Uncertainty: 0.0000048
Coverage Factor: 2

Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003

M_{234U}: Type B normal distribution
Value: 234.0409504
Expanded Uncertainty: 0.0000024
Coverage Factor: 2.0

Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003

M_{235U}: Type B normal distribution
Value: 235.0439282
Expanded Uncertainty: 0.0000024
Coverage Factor: 2.0

Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003

M_{236U}: Type B normal distribution
Value: 236.0455662
Expanded Uncertainty: 0.0000024
Coverage Factor: 2.0

Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003

M_{238U}: Type B normal distribution
Value: 238.0507870
Expanded Uncertainty: 0.0000032
Coverage Factor: 2.0

Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003

Interim Results:

Quantity	Value	Standard Uncertainty
ΣR_U	1.2542784	$20.4 \cdot 10^{-6}$

Results:

Quantity	Value	Expanded Uncertainty	Coverage factor	Coverage
M_U	237.441214	$78 \cdot 10^{-6}$	2.00	manual
f_{234U}	$3.931 \cdot 10^{-6}$	$28 \cdot 10^{-9}$	2.00	manual
f_{235U}	0.202717	$26 \cdot 10^{-6}$	2.00	manual
f_{236U}	$7.818 \cdot 10^{-6}$	$50 \cdot 10^{-9}$	2.00	manual
f_{238U}	0.797271	$26 \cdot 10^{-6}$	2.00	manual
w_{234U}	$3.875 \cdot 10^{-6}$	$27 \cdot 10^{-9}$	2.00	manual
w_{235U}	0.200670	$26 \cdot 10^{-6}$	2.00	manual
w_{236U}	$7.772 \cdot 10^{-6}$	$50 \cdot 10^{-9}$	2.00	manual
w_{238U}	0.799318	$26 \cdot 10^{-6}$	2.00	manual

Annex 5. Characterization of the minor isotope ratios for IRMM-3035

IRMM-3035

Original folder: G:\JRC.G.2\Nuclear Safeguards\Nuclear\TRITON DATA - SHARED\IRMM-3000 Series Characterization

File Folder: U:\Nuclear Safeguards\Nuclear\IRMM-3000 Series - Certification Folder\Characterisation assessment \COPY of IRMM-3000 Series Characterization

For 235U/238U ratios: T190705 - IRMM-3035-3050 Turret 5B - F-D-00447 Revision 3.xls

For 234U/238U and 236U/238U ratios: T190613 MTE - IRMM-3035-3050 - Turret-2B - normalized to DS.xls

Model Equation:

$f_{233U} = R_{233/238U}/\Sigma R_U;$

$f_{234U} = R_{234/238U}/\Sigma R_U;$

$f_{235U} = R_{235/238U}/\Sigma R_U;$

$f_{236U} = R_{236/238U}/\Sigma R_U;$

$f_{238U} = 1/\Sigma R_U;$

$\Sigma R_U = R_{233/238U} + R_{234/238U} + R_{235/238U} + R_{236/238U} + 1;$

$M_U = M_{233U}\cdot f_{233U} + M_{234U}\cdot f_{234U} + M_{235U}\cdot f_{235U} + M_{236U}\cdot f_{236U} + M_{238U}\cdot f_{238U};$

$w_{233U} = f_{233U}\cdot M_{233U}/M_U;$

$w_{234U} = f_{234U}\cdot M_{234U}/M_U;$

$w_{235U} = f_{235U}\cdot M_{235U}/M_U;$

$w_{236U} = f_{236U}\cdot M_{236U}/M_U;$

$w_{238U} = f_{238U}\cdot M_{238U}/M_U;$

List of Quantities:

Quantity	Unit	Definition
$R_{233/238U}$		isotope amount ratio n_{233}/n_{238} of U
$R_{234/238U}$		isotope amount ratio n_{234}/n_{238} of U
$R_{235/238U}$		isotope amount ratio n_{235}/n_{238} of U
$R_{236/238U}$		isotope amount ratio n_{236}/n_{238} of U
M_U		molar mass of U
f_{233U}		isotope amount fraction of ^{233}U in U
f_{234U}		isotope amount fraction of ^{234}U in U
f_{235U}		isotope amount fraction of ^{235}U in U
f_{236U}		isotope amount fraction of ^{236}U in U
f_{238U}		isotope amount fraction of ^{238}U in U
w_{233U}		isotope mass fraction of ^{233}U in U
w_{234U}		isotope mass fraction of ^{234}U in U
w_{235U}		isotope mass fraction of ^{235}U in U
w_{236U}		isotope mass fraction of ^{236}U in U
w_{238U}		isotope mass fraction of ^{238}U in U
M_{233U}		atomic mass for ^{233}U

Quantity	Unit	Definition
M_{234U}		atomic mass for ^{234}U
M_{235U}		atomic mass for ^{235}U
M_{236U}		atomic mass for ^{236}U
M_{238U}		atomic mass for ^{238}U
ΣR_U		sum of isotope ratios for U

$R_{233/238U}$:
Type B normal distribution
Value: 0
Expanded Uncertainty: 0
Coverage Factor: 2

$R_{234/238U}$:
Import from Excel
Filename: T190613 MTE - IRMM-3035-3050 - Turret-2B - 3035 norm DS 3050 norm GRAV.xls
Worksheet: Summary-calc
Value Cell: H179 = $10.6691 \cdot 10^{-6}$
Standarduncertainty Cell: H180 = $34.6 \cdot 10^{-9}$
Degrees of Freedom Cell: G51 = 5

$R_{235/238U}$:
Import from Excel
Filename: T190705 - IRMM-3035-3050 Turret 5B - F-D-00447 Revision 3.xls
Worksheet: DS-Summary
Value Cell: C73 = 0.5485313
Standarduncertainty Cell: C78 = $44.5 \cdot 10^{-6}$
Degrees of Freedom Cell: A75 = 6

$R_{236/238U}$:
Import from Excel
Filename: T190613 MTE - IRMM-3035-3050 - Turret-2B - 3035 norm DS 3050 norm GRAV.xls
Worksheet: Summary-calc
Value Cell: I179 = $21.2240 \cdot 10^{-6}$
Standarduncertainty Cell: I180 = $43.2 \cdot 10^{-9}$
Degrees of Freedom Cell: G51 = 5

M_{233U}:
Type B normal distribution
Value: 233.0396344
Expanded Uncertainty: 0.0000048
Coverage Factor: 2

Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003

M_{234U}:
Type B normal distribution
Value: 234.0409504
Expanded Uncertainty: 0.0000024
Coverage Factor: 2.0

Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003

M_{235U}:
Type B normal distribution
Value: 235.0439282
Expanded Uncertainty: 0.0000024
Coverage Factor: 2.0

Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003

M_{236U}: Type B normal distribution
Value: 236.0455662
Expanded Uncertainty: 0.0000024
Coverage Factor: 2.0

Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003

M_{238U}: Type B normal distribution
Value: 238.0507870
Expanded Uncertainty: 0.0000032
Coverage Factor: 2.0

Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003

Interim Results:

Quantity	Value	Standard Uncertainty
ΣR_U	1.5485632	$44.5 \cdot 10^{-6}$

Results:

Quantity	Value	Expanded Uncertainty	Coverage factor	Coverage
M_U	236.98564	$110 \cdot 10^{-6}$	2.00	manual
f_{234U}	$6.890 \cdot 10^{-6}$	$45 \cdot 10^{-9}$	2.00	manual
f_{235U}	0.354220	$37 \cdot 10^{-6}$	2.00	manual
f_{236U}	$13.706 \cdot 10^{-6}$	$56 \cdot 10^{-9}$	2.00	manual
f_{238U}	0.645760	$37 \cdot 10^{-6}$	2.00	manual
w_{234U}	$6.804 \cdot 10^{-6}$	$44 \cdot 10^{-9}$	2.00	manual
w_{235U}	0.351317	$37 \cdot 10^{-6}$	2.00	manual
w_{236U}	$13.651 \cdot 10^{-6}$	$56 \cdot 10^{-9}$	2.00	manual
w_{238U}	0.648662	$37 \cdot 10^{-6}$	2.00	manual

Annex 6. Characterization of the minor isotope ratios for IRMM-3050

IRMM-3050

Original folder: G:\JRC.G.2\Nuclear Safeguards\Nuclear\TRITON DATA - SHARED\IRMM-3000 Series Characterization

For 235U/238U ratios: U:\Nuclear Safeguards\Nuclear\IRMM-3000 Series - Certification Folder\Characterisation assessment\Gravimetrical Calculations \Gravimetric Calc of Ratios\IRMM-3035 Mixing NEW IMP after KA.smu

For 234U/238U and 236U/238U ratios: U:\Nuclear Safeguards\Nuclear\IRMM-3000 Series - Certification Folder \Characterisation assessment\COPY of IRMM-3000 Series Characterization\T190613 MTE - IRMM-3035-3050 - Turret-2B - 3050 normalized to GRAV

Model Equation:

$f_{233U} = R_{233/238U}/\Sigma R_U;$

$f_{234U} = R_{234/238U}/\Sigma R_U;$

$f_{235U} = R_{235/238U}/\Sigma R_U;$

$f_{236U} = R_{236/238U}/\Sigma R_U;$

$f_{238U} = 1/\Sigma R_U;$

$\Sigma R_U = R_{233/238U} + R_{234/238U} + R_{235/238U} + R_{236/238U} + 1;$

$M_U = M_{233U} \cdot f_{233U} + M_{234U} \cdot f_{234U} + M_{235U} \cdot f_{235U} + M_{236U} \cdot f_{236U} + M_{238U} \cdot f_{238U};$

$w_{233U} = f_{233U} \cdot M_{233U}/M_U;$

$w_{234U} = f_{234U} \cdot M_{234U}/M_U;$

$w_{235U} = f_{235U} \cdot M_{235U}/M_U;$

$w_{236U} = f_{236U} \cdot M_{236U}/M_U;$

$w_{238U} = f_{238U} \cdot M_{238U}/M_U;$

List of Quantities:

Quantity	Unit	Definition
$R_{233/238U}$	mol/mol	isotope amount ratio n_{233}/n_{238} of U
$R_{234/238U}$	mol/mol	isotope amount ratio n_{234}/n_{238} of U
$R_{235/238U}$	mol/mol	isotope amount ratio n_{235}/n_{238} of U
$R_{236/238U}$	mol/mol	isotope amount ratio n_{236}/n_{238} of U
M_U		molar mass of U
f_{233U}		isotope amount fraction of ^{233}U in U
f_{234U}		isotope amount fraction of ^{234}U in U
f_{235U}		isotope amount fraction of ^{235}U in U
f_{236U}		isotope amount fraction of ^{236}U in U
f_{238U}		isotope amount fraction of ^{238}U in U
w_{233U}		isotope mass fraction of ^{233}U in U
w_{234U}		isotope mass fraction of ^{234}U in U
w_{235U}		isotope mass fraction of ^{235}U in U
w_{236U}		isotope mass fraction of ^{236}U in U
w_{238U}		isotope mass fraction of ^{238}U in U

Quantity	Unit	Definition
M_{233U}		atomic mass for ^{233}U
M_{234U}		atomic mass for ^{234}U
M_{235U}		atomic mass for ^{235}U
M_{236U}		atomic mass for ^{236}U
M_{238U}		atomic mass for ^{238}U
ΣR_U		sum of isotope ratios for U

$R_{233/238U}$:
Type B rectangular distribution
Value: 0 mol/mol
Halfwidth of Limits: 0 mol/mol

$R_{234/238U}$:
Import from Excel
Filename: T190613 MTE - IRMM-3035-3050 - Turret-2B - 3035 norm DS 3050 norm GRAV.xls
Worksheet: Summary-calc
Value Cell: C192 = $19.9089 \cdot 10^{-6}$ mol/mol
Standarduncertainty Cell: C193 = $63.7 \cdot 10^{-9}$ mol/mol
Degrees of Freedom Cell: G70 = 5

$R_{235/238U}$:
Import
Filename: ..\Gravimetrical Calculations\Gravimetric Calc of Ratios\IRMM-3050 Mixing NEW IMP after KA.smu
Symbol: $R_{235/238U}$

$R_{236/238U}$:
Import from Excel
Filename: T190613 MTE - IRMM-3035-3050 - Turret-2B - 3035 norm DS 3050 norm GRAV.xls
Worksheet: Summary-calc
Value Cell: D192 = $39.6931 \cdot 10^{-6}$ mol/mol
Standarduncertainty Cell: D193 = $60.8 \cdot 10^{-9}$ mol/mol
Degrees of Freedom Cell: G70 = 5

M_{233U}:
Type B normal distribution
Value: 233.0396344
Expanded Uncertainty: 0.0000048
Coverage Factor: 2

The mass of the isotope is read from \\Sim_server\gumwork\wizards

M_{234U}:
Type B normal distribution
Value: 234.0409504
Expanded Uncertainty: 0.0000024
Coverage Factor: 2.0

Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003

M_{235U}:
Type B normal distribution
Value: 235.0439282
Expanded Uncertainty: 0.0000024
Coverage Factor: 2.0

Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003

M_{236U}: Type B normal distribution
Value: 236.0455662
Expanded Uncertainty: 0.0000024
Coverage Factor: 2.0

Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003

M_{238U}: Type B normal distribution
Value: 238.0507870
Expanded Uncertainty: 0.0000032
Coverage Factor: 2.0

Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003

Interim Results:

Quantity	Value	Standard Uncertainty
ΣR_U	2.0236482	$89.5\cdot10^{-6}$

Results:

Quantity	Value	Expanded Uncertainty	Coverage factor	Coverage
M_U	236.52980	$130\cdot10^{-6}$	2.00	manual
f_{234U}	$9.838\cdot10^{-6}$	$63\cdot10^{-9}$	2.00	manual
f_{235U}	0.505814	$44\cdot10^{-6}$	2.00	manual
f_{236U}	$19.615\cdot10^{-6}$	$60\cdot10^{-9}$	2.00	manual
f_{238U}	0.494157	$44\cdot10^{-6}$	2.00	manual
w_{234U}	$9.735\cdot10^{-6}$	$62\cdot10^{-9}$	2.00	manual
w_{235U}	0.502636	$44\cdot10^{-6}$	2.00	manual
w_{236U}	$19.574\cdot10^{-6}$	$60\cdot10^{-9}$	2.00	manual
w_{238U}	0.497335	$44\cdot10^{-6}$	2.00	manual

Annex 7. Characterization of the minor isotope ratios for IRMM-3075

IRMM-3075

Original folder: G:\JRC.G.2\Nuclear Safeguards\Nuclear\TRITON DATA - SHARED\IRMM-3000 Series Characterization

For 235U/238U ratios: U:\Nuclear Safeguards\Nuclear\IRMM-3000 Series - Certification Folder\Characterisation assessment \Gravimetrical Calculations \Gravimetric Calc of Ratios\IRMM-3075 Mixing NEW IMP after KA.smu

For 234U/238U and 236U/238U ratios: U:\Nuclear Safeguards\Nuclear\IRMM-3000 Series - Certification Folder \Characterisation assessment\COPY of IRMM-3000 Series Characterization\T190523 MTE - IRMM-3075-3090 - Turret-3B - normalized to GRAV.xls

Model Equation:

$f_{233U} = R_{233/238U}/\Sigma R_U;$

$f_{234U} = R_{234/238U}/\Sigma R_U;$

$f_{235U} = R_{235/238U}/\Sigma R_U;$

$f_{236U} = R_{236/238U}/\Sigma R_U;$

$f_{238U} = 1/\Sigma R_U;$

$\Sigma R_U = R_{233/238U} + R_{234/238U} + R_{235/238U} + R_{236/238U} + 1;$

$M_U = M_{233U} \cdot f_{233U} + M_{234U} \cdot f_{234U} + M_{235U} \cdot f_{235U} + M_{236U} \cdot f_{236U} + M_{238U} \cdot f_{238U};$

$w_{233U} = f_{233U} \cdot M_{233U}/M_U;$

$w_{234U} = f_{234U} \cdot M_{234U}/M_U;$

$w_{235U} = f_{235U} \cdot M_{235U}/M_U;$

$w_{236U} = f_{236U} \cdot M_{236U}/M_U;$

$w_{238U} = f_{238U} \cdot M_{238U}/M_U;$

List of Quantities:

Quantity	Unit	Definition
$R_{233/238U}$	mol/mol	isotope amount ratio n_{233}/n_{238} of U
$R_{234/238U}$	mol/mol	isotope amount ratio n_{234}/n_{238} of U
$R_{235/238U}$	mol/mol	isotope amount ratio n_{235}/n_{238} of U
$R_{236/238U}$	mol/mol	isotope amount ratio n_{236}/n_{238} of U
M_U		molar mass of U
f_{233U}		isotope amount fraction of ^{233}U in U
f_{234U}		isotope amount fraction of ^{234}U in U
f_{235U}		isotope amount fraction of ^{235}U in U
f_{236U}		isotope amount fraction of ^{236}U in U
f_{238U}		isotope amount fraction of ^{238}U in U
w_{233U}		isotope mass fraction of ^{233}U in U
w_{234U}		isotope mass fraction of ^{234}U in U
w_{235U}		isotope mass fraction of ^{235}U in U
w_{236U}		isotope mass fraction of ^{236}U in U
w_{238U}		isotope mass fraction of ^{238}U in U

Quantity	Unit	Definition
M_{233U}		atomic mass for ^{233}U
M_{234U}		atomic mass for ^{234}U
M_{235U}		atomic mass for ^{235}U
M_{236U}		atomic mass for ^{236}U
M_{238U}		atomic mass for ^{238}U
ΣR_U		sum of isotope ratios for U

$R_{233/238U}$: Type B rectangular distribution
Value: 0 mol/mol
Halfwidth of Limits: 0 mol/mol

$R_{234/238U}$: Import from Excel
Filename: T190523 MTE - IRMM-3075-3090 - Turret-3B - normalized to GRAV.xls
Worksheet: Summary-calc
Value Cell: H179 = $54.733 \cdot 10^{-6}$ mol/mol
Standarduncertainty Cell: H180 = $174 \cdot 10^{-9}$ mol/mol
Degrees of Freedom Cell: G51 = 5

$R_{235/238U}$: Import
Filename: ..\Gravimetrical Calculations\Gravimetric Calc of Ratios\IRMM-3075 Mixing NEW IMP after KA.smu
Symbol: $R_{235/238U}$

$R_{236/238U}$: Import from Excel
Filename: T190523 MTE - IRMM-3075-3090 - Turret-3B - normalized to GRAV.xls
Worksheet: Summary-calc
Value Cell: I179 = $109.664 \cdot 10^{-6}$ mol/mol
Standarduncertainty Cell: I180 = $100 \cdot 10^{-9}$ mol/mol
Degrees of Freedom Cell: G70 = 5

M_{233U}: Type B normal distribution
Value: 233.0396344
Expanded Uncertainty: 0.0000048
Coverage Factor: 2

Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003

M_{234U}: Type B normal distribution
Value: 234.0409504
Expanded Uncertainty: 0.0000024
Coverage Factor: 2.0

Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003

M_{235U}: Type B normal distribution
Value: 235.0439282
Expanded Uncertainty: 0.0000024
Coverage Factor: 2.0

Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003

M_{236U}: Type B normal distribution
Value: 236.0455662
Expanded Uncertainty: 0.0000024
Coverage Factor: 2.0

Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003

M_{238U}: Type B normal distribution
Value: 238.0507870
Expanded Uncertainty: 0.0000032
Coverage Factor: 2.0

Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003

Interim Results:

Quantity	Value	Standard Uncertainty
ΣR_U	3.818883	$221 \cdot 10^{-6}$

Results:

Quantity	Value	Expanded Uncertainty	Coverage factor	Coverage
M_U	235.831309	$91 \cdot 10^{-6}$	2.00	manual
f_{234U}	$14.332 \cdot 10^{-6}$	$91 \cdot 10^{-9}$	2.00	manual
f_{235U}	0.738100	$30 \cdot 10^{-6}$	2.00	manual
f_{236U}	$28.716 \cdot 10^{-6}$	$53 \cdot 10^{-9}$	2.00	manual
f_{238U}	0.261857	$30 \cdot 10^{-6}$	2.00	manual
w_{234U}	$14.224 \cdot 10^{-6}$	$91 \cdot 10^{-9}$	2.00	manual
w_{235U}	0.735636	$30 \cdot 10^{-6}$	2.00	manual
w_{236U}	$28.742 \cdot 10^{-6}$	$53 \cdot 10^{-9}$	2.00	manual
w_{238U}	0.264321	$30 \cdot 10^{-6}$	2.00	manual

Annex 8. Characterization of the minor isotope ratios for IRMM-3090

IRMM-3090

Original folder: G:\JRC.G.2\Nuclear Safeguards\Nuclear\TRITON DATA - SHARED\IRMM-3000 Series Characterization

For 235U/238U ratios: U:\Nuclear Safeguards\Nuclear\IRMM-3000 Series - Certification Folder\Characterisation assessment \Gravimetrical Calculations \Gravimetric Calc of Ratios\IRMM-3090 Mixing NEW IMP after KA.smu

For 234U/238U and 236U/238U ratios: U:\Nuclear Safeguards\Nuclear\IRMM-3000 Series - Certification Folder \Characterisation assessment\COPY of IRMM-3000 Series Characterization\T190523 MTE - IRMM-3075-3090 - Turret-3B - normalized to GRAV.xls

Model Equation:

$$f_{233U} = R_{233/238U}/\Sigma R_U;$$
$$f_{234U} = R_{234/238U}/\Sigma R_U;$$
$$f_{235U} = R_{235/238U}/\Sigma R_U;$$
$$f_{236U} = R_{236/238U}/\Sigma R_U;$$
$$f_{238U} = 1/\Sigma R_U;$$
$$\Sigma R_U = R_{233/238U} + R_{234/238U} + R_{235/238U} + R_{236/238U} + 1;$$

$$M_U = M_{233U} \cdot f_{233U} + M_{234U} \cdot f_{234U} + M_{235U} \cdot f_{235U} + M_{236U} \cdot f_{236U} + M_{238U} \cdot f_{238U};$$
$$w_{233U} = f_{233U} \cdot M_{233U}/M_U;$$
$$w_{234U} = f_{234U} \cdot M_{234U}/M_U;$$
$$w_{235U} = f_{235U} \cdot M_{235U}/M_U;$$
$$w_{236U} = f_{236U} \cdot M_{236U}/M_U;$$
$$w_{238U} = f_{238U} \cdot M_{238U}/M_U;$$

List of Quantities:

Quantity	Unit	Definition
$R_{233/238U}$	mol/mol	isotope amount ratio n_{233}/n_{238} of U
$R_{234/238U}$	mol/mol	isotope amount ratio n_{234}/n_{238} of U
$R_{235/238U}$	mol/mol	isotope amount ratio n_{235}/n_{238} of U
$R_{236/238U}$	mol/mol	isotope amount ratio n_{236}/n_{238} of U
M_U		molar mass of U
f_{233U}		isotope amount fraction of ^{233}U in U
f_{234U}		isotope amount fraction of ^{234}U in U
f_{235U}		isotope amount fraction of ^{235}U in U
f_{236U}		isotope amount fraction of ^{236}U in U
f_{238U}		isotope amount fraction of ^{238}U in U
w_{233U}		isotope mass fraction of ^{233}U in U
w_{234U}		isotope mass fraction of ^{234}U in U
w_{235U}		isotope mass fraction of ^{235}U in U
w_{236U}		isotope mass fraction of ^{236}U in U
w_{238U}		isotope mass fraction of ^{238}U in U

Quantity	Unit	Definition
M_{233U}		atomic mass for ^{233}U
M_{234U}		atomic mass for ^{234}U
M_{235U}		atomic mass for ^{235}U
M_{236U}		atomic mass for ^{236}U
M_{238U}		atomic mass for ^{238}U
ΣR_U		sum of isotope ratios for U

$R_{233/238U}$: Type B rectangular distribution
Value: 0 mol/mol
Halfwidth of Limits: 0 mol/mol

$R_{234/238U}$: Import from Excel
Filename: T190523 MTE - IRMM-3075-3090 - Turret-3B - normalized to GRAV.xls
Worksheet: Summary-calc
Value Cell: C192 = $127.700 \cdot 10^{-6}$ mol/mol
Standarduncertainty Cell: C193 = $413 \cdot 10^{-9}$ mol/mol
Degrees of Freedom Cell: G70 = 5

$R_{235/238U}$: Import
Filename: ..\Gravimetrical Calculations\Gravimetric Calc of Ratios\IRMM-3090 Mixing NEW IMP after KA.smu
Symbol: $R_{235/238U}$

$R_{236/238U}$: Import from Excel
Filename: T190523 MTE - IRMM-3075-3090 - Turret-3B - normalized to GRAV.xls
Worksheet: Summary-calc
Value Cell: D192 = $256.536 \cdot 10^{-6}$ mol/mol
Standarduncertainty Cell: D193 = $172 \cdot 10^{-9}$ mol/mol
Degrees of Freedom Cell: G70 = 5

M_{233U}: Type B normal distribution
Value: 233.0396344
Expanded Uncertainty: 0.0000048
Coverage Factor: 2

Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003

M_{234U}: Type B normal distribution
Value: 234.0409504
Expanded Uncertainty: 0.0000024
Coverage Factor: 2.0

Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003

M_{235U}: Type B normal distribution
Value: 235.0439282
Expanded Uncertainty: 0.0000024
Coverage Factor: 2.0

Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003

M_{236U}: Type B normal distribution
Value: 236.0455662
Expanded Uncertainty: 0.0000024
Coverage Factor: 2.0

Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003

M_{238U}: Type B normal distribution
Value: 238.0507870
Expanded Uncertainty: 0.0000032
Coverage Factor: 2.0

Wang et al., The AME 2016 atomic mass evaluation, Chinese Physics C Vol. 41, No. 3 (2017) 030003

Interim Results:

Quantity	Value	Standard Uncertainty
ΣR_U	7.577697	$461 \cdot 10^{-6}$

Results:

Quantity	Value	Expanded Uncertainty	Coverage factor	Coverage
M_U	235.440749	$48 \cdot 10^{-6}$	2.00	manual
f_{234U}	$16.85 \cdot 10^{-6}$	$110 \cdot 10^{-9}$	2.00	manual
f_{235U}	0.867983	$16 \cdot 10^{-6}$	2.00	manual
f_{236U}	$33.854 \cdot 10^{-6}$	$46 \cdot 10^{-9}$	2.00	manual
f_{238U}	0.131966	$16 \cdot 10^{-6}$	2.00	manual
w_{234U}	$16.75 \cdot 10^{-6}$	$110 \cdot 10^{-9}$	2.00	manual
w_{235U}	0.866520	$16 \cdot 10^{-6}$	2.00	manual
w_{236U}	$33.941 \cdot 10^{-6}$	$46 \cdot 10^{-9}$	2.00	manual
w_{238U}	0.133429	$16 \cdot 10^{-6}$	2.00	manual

Annex 9. Peak tailing correction for the minor ratios and associated uncertainties

In this annex, the uncertainties for the peak tailing correction caused by the uncertainties originating from the mass calibration curve, i.e. the calibration of the detected mass versus the applied magnet current, are evaluated. The uncertainties from the mass calibration can be obtained from the recordings of the peak-centring actions, which are written into the logfiles during the measurements. Typical values for the mass uncertainties are at the level of 0.01-0.03 amu. For the measurement of the ion beams for the isotopes ^{234}U, ^{235}U, ^{236}U and ^{238}U this has no effect because of the flat peak tops of the Faraday detectors, which are due to the ion beam widths being smaller than the widths of the Faraday cups. But for the measurements of the peak tail corrections, the ion beams at the masses 233.7, 234.4, 234.7, 235.4, 235.7, 236.4, 237.7 and 238.4 have to be measured (≈±0.35 amu below and beyond the uranium isotope masses ^{234}U, ^{235}U, ^{236}U and ^{238}U), and for these masses the tailing curve, i.e. the ion beam versus mass function, is not flat. The curve might be associated with a significant slope, which causes uncertainties depending on the uncertainty of the mass setting and mass calibration.

Apart from statistical variations of the mass calibration, there could also be a continuous drift over time, possibly due to a drift in the ambient conditions. In addition, the selection of the isotope masses included in the mass calibration table and the frequency of updating the calibration is different among the involved laboratories, JRC-G.2, ORNL and the IAEA. As a conservative estimate, an uncertainty value of 0.03 amu (k=2) for each mass setting is taken into account.

The value for the peak tailing correction of the minor isotope ratios $^{234}U/^{238}U$ has to be interpolated between the measured ratios $^{233.7}U/^{238}U$ and $^{234.4}U/^{238}U$. Similarly, the $^{236}U/^{238}U$ correction value is an interpolation between $^{235.7}U/^{238}U$ and $^{236.4}U/^{238}U$. There are several ways the interpolation can be calculated, mainly using a geometric mean or an arithmetic mean. The geometric mean is calculated assuming an exponential tail function fitted through the two tail measurements taken. The geometric mean can also be considered as the arithmetic mean, i.e. the average of two values on a linear regression line, in the logarithmic scale.

Exponential interpolation, geometric mean:

The exponential tailing function of $E(m)$ at mass m is given by

$$E(m) = a\,e^{bm} \quad \text{eq (1)}$$

The slope of the tailing versus the mass m is

$$\frac{dE(m)}{dm} = b \times\ a\,e^{bm} = b \times E(m) \quad \text{eq (2)}$$

This function has 2 parameters, a and b.

Equation (1) can be applied to masses m_1 and m_2 on the low and high mass side of m:

$$E(m_1) = a\,e^{bm_1} \quad \text{eq (3)}$$

$$E(m_2) = a\,e^{bm_2} \quad \text{eq (4)}$$

Using these 2 equations, the parameters a and b can be derived:

$$b = \ln\left(\frac{E(m_1)}{E(m_2)}\right) \frac{1}{m_1 - m_2} \quad \text{eq (5)}$$

$$a = \frac{E(m_2)}{e^{bm_2}} \quad \text{eq (6)}$$

This is the same as

$$a = \frac{E(m_1)}{e^{bm_1}} \quad \text{eq (7)}$$

The tailing can be interpolated at the mass $(m_1+m_2)/2$ between m_1 and m_2, leading to the so-called "geometric mean":

$$E((m_1 + m_2)/2) = a\,e^{\frac{b(m_1+m_2)}{2}} = \sqrt{\left(E(m_1) \times E(m_2)\right)} \quad \text{eq (8)}$$

For the uncertainty calculations, the partial derivatives of the geometric mean $E((m_1 + m_2)/2)$ versus m_1 and m_2 have to be calculated:

$$\frac{\partial E((m_1+m_2)/2)}{\partial m_1} = \frac{b \times E(m_1) \times E(m_2)}{2 \times \sqrt{(E(m_1) \times E(m_2))}} \qquad \text{eq (9)}$$

$$\frac{\partial E((m_1+m_2)/2)}{\partial m_2} = \frac{b \times E(m_1) \times E(m_2)}{2 \times \sqrt{(E(m_1) \times E(m_2))}} \qquad \text{eq (10)}$$

The combined uncertainty of the geometric mean $E((m_1 + m_2)/2)$ can be calculated using the error propagation law:

$$\Delta E((m_1 + m_2)/2) = \frac{|b|}{2} \sqrt{(E(m_1) \times E(m_2))} \times \sqrt{(\Delta m_1)^2 + (\Delta m_2)^2} \qquad \text{eq (11)}$$

Assuming that the two independent masses m_1 and m_2 have the same values for their uncertainties Δm_1 and Δm_2, the mass uncertainty can be expressed as $\Delta m_{1,2}$, leading to

$$\Delta E((m_1 + m_2)/2) = \frac{|b|}{\sqrt{2}} \sqrt{(E(m_1) \times E(m_2))} \times \Delta m_{1,2} \qquad \text{eq (12)}$$

By entering the result for b from eq (5) this results in

$$\Delta E((m_1 + m_2)/2) = \left| \ln\left(\frac{E(m_1)}{E(m_2)}\right) \frac{1}{m_1 - m_2} \right| \frac{1}{\sqrt{2}} \sqrt{(E(m_1) \times E(m_2))} \times \Delta m_{1,2} \qquad \text{eq (13)}$$

Arithmetic Interpolation:

For an arithmetic interpolation, the tailing function of *A(m)* at mass *m* is given by

$$A(m) = a + b \times m \qquad \text{eq (14)}$$

The slope of the tailing versus the mass m is

$$\frac{dA(m)}{dm} = b \qquad \text{eq (15)}$$

This function has 2 parameters, a and b.

Applying eq. (14) to masses m_1 and m_2 on the low and high mass side of *m,* leads to:

$$A(m_1) = a + b \times m_1 \qquad \text{eq (16)}$$

$$A(m_2) = a + b \times m_2 \qquad \text{eq (17)}$$

The measured numerical values $A(m_1), and\ A(m_2)$ are the same as $E(m_1), and\ E(m_2)$, but the functions have been given different symbols for clarifying the different ways of extrapolation.

Using these 2 equations, the parameters a and b can be derived:

$$b = \frac{A(m_1) - A(m_2)}{m_1 - m_2} \qquad \text{eq (18)}$$

$$a = A(m_1) - m_1 \times \frac{A(m_1) - A(m_2)}{m_1 - m_2} \qquad \text{eq (19)}$$

This is the same as

$$a = A(m_2) - m_2 \times \frac{A(m_1) - A(m_2)}{m_1 - m_2} \qquad \text{eq (20)}$$

The tailing can be interpolated at the mass between m_1 and m_2 , leading to the so-called “arithmetic mean”:

$$A((m_1 + m_2)/2) = \left(A(m_1) + A(m_2)\right)/2 \qquad \text{eq (21)}$$

For the uncertainty calculations, the partial derivatives of $A((m_1 + m_2)/2)$ versus m_1 and m_2 have to be calculated:

$$\frac{\partial A((m_1+m_2)/2)}{\partial m_1} = \frac{b}{2} \qquad \text{eq (22)}$$

$$\frac{\partial A((m_1+m_2)/2)}{\partial m_2} = \frac{b}{2} \qquad \text{eq (23)}$$

The combined uncertainty of the arithmetic mean $A((m_1 + m_2)/2)$ can be calculated using the error propagation law:

$$\Delta A((m_1 + m_2)/2) = \frac{|b|}{2} \times \sqrt{(\Delta m_1)^2 + (\Delta m_2)^2} \qquad \text{eq (24)}$$

Assuming that the independent masses m_1 and m_2 have the same values for the uncertainties Δm_1 and Δm_2, the mass uncertainty can be expressed as $\Delta m_{1,2}$, leading to

$$\Delta A((m_1 + m_2)/2) = \frac{|b|}{\sqrt{2}} \times \Delta m_{1,2} \qquad \text{eq (25)}$$

By entering the result for b from eq (21) this results in

$$\Delta A((m_1 + m_2)/2) = \left|\frac{A(m_1)-A(m_2)}{m_1-m_2}\right| \frac{1}{\sqrt{2}} \times \Delta m_{1,2} \qquad \text{eq (26)}$$

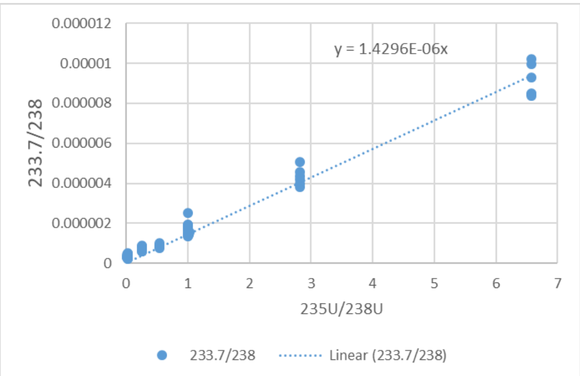
Results for tailing uncertainties based on mass uncertainties

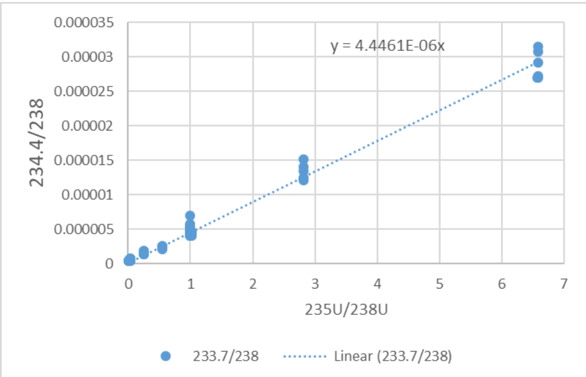
As a conclusion, eq(13) and eq(26) can be used to calculate the uncertainties $\Delta E((m_1 + m_2)/2)$ and $\Delta A((m_1 + m_2)/2)$ of the tailing correction, using the known values for the masses m_1, m_2, and the measured tailing ratios $E(m_1)$, $E(m_2)$, which have the same numerical values as $A(m_1)$, $A(m_2)$.

In order to obtain quantitative values for the uncertainties $\Delta E((m_1 + m_2)/2)$ and $\Delta A((m_1 + m_2)/2)$ of the tailing correction, all tailing ratios $E(m_1)$ and $E(m_2)$ measured during the IRMM-3000 project have been collected for a detailed evaluation. This collection includes data from the reference material IRMM-074 measured for mass fractionation correction during MTE analyses, the QC samples IRMM-075 and IRMM-2023 and the IRMM-3000 series materials IRMM-3020, IRMM-3035, IRMM-3050, IRMM-3075 and IRMM-3090 themselves, all together covering a total ^{235}U enrichment range of 0.7% to 90%.

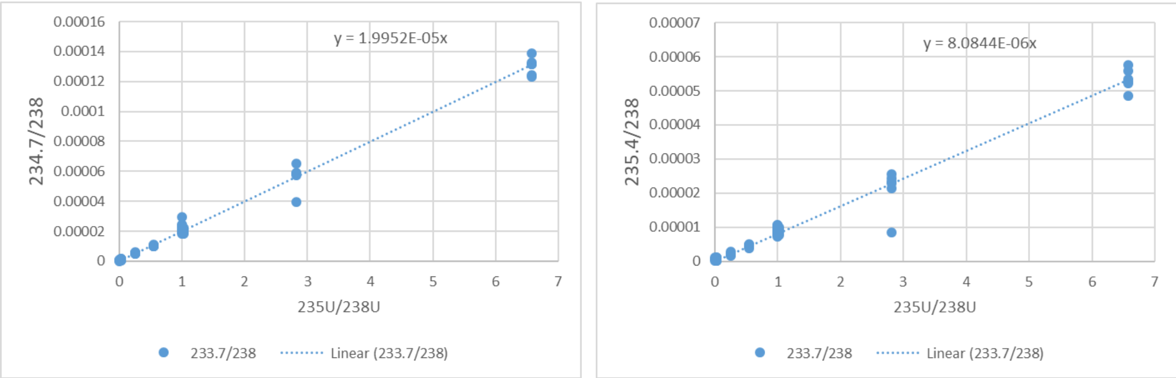
The measured tailing ratios $n(^{233.7}U)/n(^{238}U)$, $n(^{234.4}U)/n(^{238}U)$, $n(^{234.7}U)/n(^{238}U)$, $n(^{235.4}U)/n(^{238}U)$, $n(^{235.7}U)/n(^{238}U)$, $n(^{236.4}U)/n(^{238}U)$, $n(^{237.7}U)/n(^{238}U)$ and $n(^{238.4}U)/n(^{238}U)$ are plotted versus the major ratios $n(^{235}U)/n(^{238}U)$ in Figures A9.1a-h below. It can be observed that the tailing ratios $n(^{233.7}U)/n(^{238}U)$, $n(^{234.4}U)/n(^{238}U)$, $n(^{234.7}U)/n(^{238}U)$, $n(^{235.4}U)/n(^{238}U)$, $n(^{235.7}U)/n(^{238}U)$ are directly proportional to the $n(^{235}U)/n(^{238}U)$ ratios. For $n(^{236.4}U)/n(^{238}U)$, there is no proportionality anymore but at least a linear relationship with the $n(^{235}U)/n(^{238}U)$ ratios, with an intercept different from zero. This is due to the fact that even for the lowest $n(^{235}U)/n(^{238}U)$ ratios for natural samples, e.g. for IRMM-075 which is a mixture of natural uranium with ^{236}U, a significant tailing contribution also from the ^{238}U ion beam is observed. For the tailing ratios $n(^{237.7}U)/n(^{238}U)$ and $n(^{238.4}U)/n(^{238}U)$ there is no influence from the ^{235}U ion beam any more, and that is the reason why no slope but rather constant tailing ratios are observed.

Figures A9.1a,b: measured ratios $n(^{233.7}U)/n(^{238}U)$, $n(^{234.4}U)/n(^{238}U)$ plotted versus $n(^{235}U)/n(^{238}U)$.

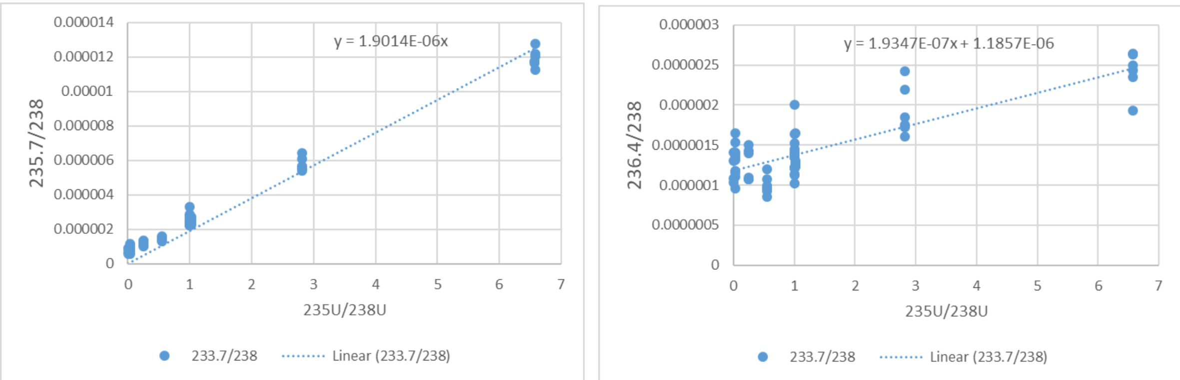




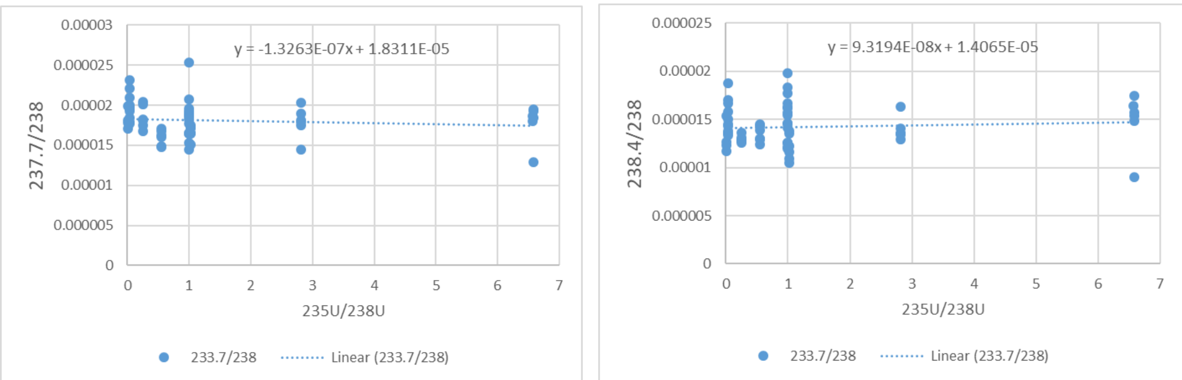
Figures A9.1c,d: measured ratios $n(^{234.7}\mathrm{U})/n(^{238}\mathrm{U})$, $n(^{235.4}\mathrm{U})/n(^{238}\mathrm{U})$ plotted versus $n(^{235}\mathrm{U})/n(^{238}\mathrm{U})$.



Figures A9.1e,f: measured ratios $n(^{235.7}\mathrm{U})/n(^{238}\mathrm{U})$, $n(^{236.4}\mathrm{U})/n(^{238}\mathrm{U})$ plotted versus $n(^{235}\mathrm{U})/n(^{238}\mathrm{U})$.



Figures A9.1g,h: measured ratios $n(^{234.7}\mathrm{U})/n(^{238}\mathrm{U})$, $n(^{235.4}\mathrm{U})/n(^{238}\mathrm{U})$ plotted versus $n(^{235}\mathrm{U})/n(^{238}\mathrm{U})$.



The tailing data shown in Figures A9.1a-h, including the calculated slopes and intercepts (where applicable), can be used to calculate the tailing ratios for each value of the major ratio $n(^{235}\mathrm{U})/n(^{238}\mathrm{U})$, as well as the uncertainties $\Delta E((m_1 + m_2)/2)$ and $\Delta A((m_1 + m_2)/2)$ of the tailing corrections according to eq(13) and eq(26). In Tables A9.1 and A9.2 the observed tailing ratios and the uncertainties of the tailing corrections for $n(^{234}\mathrm{U})/n(^{238}\mathrm{U})$ and $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ ratios are shown for both the arithmetic and the geometric mean, in absolute numbers and relative to the ratios. The uncertainties are expressed with a coverage factor k=2, based on a mass uncertainty of 0.03 amu (k=2).

Between the two ways of interpolation, using the arithmetic or the geometric mean, there is not much of a difference in the tailing uncertainties. For the $n(^{234}\mathrm{U})/n(^{238}\mathrm{U})$ ratios for the LEU materials IRMM-075/1 (close to natural uranium, ≈0.7 % ^{235}U) and IRMM-2023 (≈3 % ^{235}U), it is obvious that the relative uncertainties from the tailing corrections are quite small, they are usually negligible.

Table A9.1 Tailing uncertainties (k=2) for $n(^{234}U)/n(^{238}U)$ ratios, based on a mass uncertainty of 0.03 amu (k=2).

Material	$n(^{235}U)/n(^{238}U)$	$n(^{234}U)/n(^{238}U)$	Measured tailing $n(^{233.7}U)/n(^{238}U)$	Measured tailing $n(^{234.4}U)/n(^{238}U)$	Arithm. Mean, Absolute Tailing Uncertainty (k=2), eq(13)	Arithm. Mean, Tailing Unc., rel to ratio (k=2)	Geom. Mean, Absolute Tailing Uncertainty (k=2), eq(26)	Geom. Mean, Tailing Unc., rel to ratio (k=2)
IRMM-075/1	0.00726	0.0000533	1.04E-08	3.23E-08	6.64E-10	0.0012%	6.29E-10	0.0012%
IRMM-2023	0.03388	0.0003396	4.84E-08	1.51E-07	3.10E-09	0.00091%	2.94E-09	0.00087%
IRMM-3020	0.25426	0.0000049	3.63E-07	1.13E-06	2.32E-08	0.47%	2.20E-08	0.45%
IRMM-3035	0.54853	0.0000107	7.84E-07	2.44E-06	5.02E-08	0.47%	4.76E-08	0.45%
IRMM-3050	1.02359	0.0000199	1.46E-06	4.55E-06	9.36E-08	0.47%	8.87E-08	0.45%
IRMM-3075	2.81872	0.0000547	4.03E-06	1.25E-05	2.58E-07	0.47%	2.44E-07	0.45%
IRMM-3090	6.57731	0.0001276	9.40E-06	2.92E-05	6.01E-07	0.47%	5.70E-07	0.45%

Table A9.2 Tailing uncertainties (k=2) for $n(^{236}U)/n(^{238}U)$ ratios, based on a mass uncertainty of 0.03 amu (k=2).

Material	$n(^{235}U)/n(^{238}U)$	$n(^{236}U)/n(^{238}U)$	Measured tailing $n(^{235.7}U)/n(^{238}U)$	Measured tailing $n(^{236.4}U)/n(^{238}U)$	Arithm. Mean, Absolute Tailing Uncertainty (k=2), eq(13)	Arithm. Mean, Tailing Unc., rel to ratio (k=2)	Geom. Mean, Absolute Tailing Uncertainty (k=2), eq(26)	Geom. Mean, Tailing Unc., rel to ratio (k=2)
IRMM-075/1	0.0072603	0.0001044	1.38E-08	1.19E-06	3.56E-08	0.034%	1.73E-08	0.017%
IRMM-2023	0.0338814	0.0000001	6.44E-08	1.19E-06	3.42E-08	29.6%	2.45E-08	21.3%
IRMM-3020	0.25426	0.0000098	4.83E-07	1.23E-06	2.28E-08	0.23%	2.20E-08	0.22%
IRMM-3035	0.54853	0.0000212	1.04E-06	1.29E-06	7.54E-09	0.036%	7.53E-09	0.036%
IRMM-3050	1.02359	0.0000396	1.95E-06	1.38E-06	1.70E-08	0.043%	1.70E-08	0.043%
IRMM-3075	2.81872	0.0001095	5.36E-06	1.73E-06	1.10E-07	0.10%	1.04E-07	0.10%
IRMM-3090	6.57731	0.0002563	1.25E-05	2.46E-06	3.04E-07	0.12%	2.73E-07	0.11%

For HEU materials the tailing uncertainties are much larger. For example, for IRMM-3090 the tailing on the low and high mass sides of ^{234}U as expressed by the ratios $n(^{233.7}U)/n(^{238}U)$ and $n(^{234.4}U)/n(^{238}U)$ can be as large as 9.4×10^{-6} and 2.92×10^{-5}, respectively, which is about 7.4 % and 23 %, respectively, of the measured $n(^{234}U)/n(^{238}U)$ ratio of 0.0001276. The uncertainties (k=2) of the tailing corrections are 6.01×10^{-7} and 5.70×10^{-7} for the arithmetic and geometric interpolations, respectively, which is at a level of 0.47 % and 0.45 %, respectively, relative to the measured $n(^{234}U)/n(^{238}U)$ ratio of 0.0001276.

For the entire IRMM-3000 series, the relative uncertainties from the tailing corrections are almost constant at a level of 0.45 %-0.47 % for the $n(^{234}U)/n(^{238}U)$ ratio. This is due to the fact that the ^{234}U contributions to the IRMM-3000 series are mainly originating from the ^{235}U base material. The IRMM-3000 consists of five materials with high ^{235}U enrichments of 20 % - 90 % but rather low $n(^{234}U)/n(^{238}U)$ ratios, which is the reason for this remarkable level of tailing uncertainties. This is also obvious in the uncertainty budgets shown in Table A9.3.

For the $n(^{236}U)/n(^{238}U)$ ratios, the tailing uncertainties are slightly smaller, because the tailing on the high mass side is in general, as well as in this case of the ^{235}U beam, smaller compared to the low mass side. Remarkably, for IRMM-2023 with a very low $n(^{236}U)/n(^{238}U)$ ratio, the tailing uncertainty is quite high. This is also obvious in the uncertainty budgets shown in Table A9.4.

Uncertainty budgets for minor ratios including the tailing contribution

Tables A9.3 and A9.4 show uncertainty budgets for the MTE measurements of the minor ratios $n(^{234}U)/n(^{238U})$ and $n(^{236}U)/n(^{238U})$, for the reference materials IRMM-075/1, IRMM-2023, IRMM-3050, IRMM-3075 and IRMM-3090. In case of IRMM-3050, IRMM-3075 and IRMM-3090, these budgets slightly differ from those presented in Table 10 and Table 11, which were calculated for the minor ratios normalized to the certified major ratios based on gravimetric preparation and not from the MTE measurements themselves.

As a conclusion, for HEU materials with quite low minor ratios as for the IRMM-3000 series the uncertainties for minor isotope ratio measurements can reach a level of up to 0.6 % due to the dominating uncertainty contribution from the peak tailing correction. But for HEU isotopic compositions encountered in safeguards samples and most HEU reference materials, e.g. like the NBL CRM U500, where the minor ratios are at a level of 0.001-0.05, this uncertainty contribution is not dominating. Also for depleted, natural or LEU materials, the uncertainty contribution from the peak tailing correction is usually negligible.

Table A9.3 Uncertainty budgets for the “minor” ratios $n(^{234}U)/n(^{238}U)$ measured by MTE. (*due to the rounding of the last digit, the sum of the relative uncertainty contributions can deviate by 0.1 % from the expected 100 %).

	IRMM-075/1	IRMM-2023	IRMM-3050	IRMM-3075	IRMM-3090
Ratio $n(^{234}U)/n(^{238}U)$	0.000053313	0.00033961	0.00001991	0.00005473	0.00012770
Rel. Unc. (k=2)	0.055%	0.045%	0.64%	0.64%	0.65%
Relative uncertainty contribution (*)					
Certified major ratio $n(^{235}U)/n(^{238U})$ of IRMM-074/10, for normalization	13.4%	19.5%	0.1%	0.1%	0.1%
Measured major ratio $n(^{235}U)/n(^{238U})$ of IRMM-074/10, for normalization	2.3%	3.4%	0.0%	0.1%	0.1%
Contribution from measurement of $n(^{234}U)/n(^{238U})$	43.0%	3.6%	2.8%	1.3%	3.7%
Measured major	17.3%	38.6%	0.1%	0.1%	0.1%

	IRMM-075/1	IRMM-2023	IRMM-3050	IRMM-3075	IRMM-3090
ratio $n(^{235}U)/n(^{238}U)$ of sample, for normalization					
MTE external uncertainty [9]	23.9%	34.7%	0.2%	0.2%	0.2%
Peak tailing	0.1%	0.1%	96.8%	98.3%	95.9%

Table A9.4 Uncertainty budgets for the "minor" ratios $n(^{236}U)/n(^{238}U)$ measured by MTE. (*due to the rounding of the last digit the sum of the relative uncertainty contributions can deviate by 0.1 % from the expected 100 %).

	IRMM-075/1	IRMM-2023	IRMM-3050	IRMM-3075	IRMM-3090
Ratio $n(^{236}U)/n(^{238}U)$	0.00010448	0.00000012	0.00003969	0.00010966	0.00025632
Rel. Unc. (k=2)	0.038%	55%	0.31%	0.18%	0.14%
Relative Uncertainty contribution(*):					
Certified major ratio $n(^{235}U)/n(^{238}U)$ of IRMM-074/10, for normalization	7.0%	0.0%	0.1%	0.3%	0.5%
Measured major ratio $n(^{235}U)/n(^{238}U)$ of IRMM-074/10, for normalization	1.2%	0.0%	0.0%	0.2%	0.4%
Contribution from measurement of $n(^{236}U)/n(^{238}U)$	64.1%	75.6%	12.5%	11.9%	30.9%
Measured major ratio $n(^{235}U)/n(^{238}U)$ of sample, for normalization	9.0%	0.0%	0.1%	0.2%	0.4%
MTE external uncertainty [9])	12.4%	0.0%	0.2%	0.5%	1.0%
Peak tailing	6.3%	24.4%	87.1%	86.8%	66.9%

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