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Re-Certification of the IRMM-183-187 Series of Uranium Nitrate Solution Reference Materials

Certified for Isotope Ratios

S. Richter, C. Hennessy, U. Jacobsson,
Y Aregbe, C. Hexel

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Contact information
Name: Stephan Richter
Address: Retieseweg 111, B-2440 Geel, Belgium
Email: stephan.richter@ec.europa.eu
Tel.: +32 14 571 701

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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 re-certification of the IRMM-183-187 series, a series of uranium nitrate solution isotopic reference materials with isotope amount fractions $n(^{235}U)/n(U)$ ranging from 0.3 % to 4.5 %. The project was a collaboration of JRC-G2 with the International Atomic Energy Agency (IAEA) in Seibersdorf, Austria, the Oak Ridge National Laboratory (ORNL) in Oak Ridge, USA, and the Commissariat à l'Energie Atomique et aux Energies Alternatives (CEA), GIF SUR YVETTE CEDEX, France.

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Authors:

S. Richter[1], C. Hennessy[1], U. Jacobsson[1], Y. Aregbe[1], C. Hexel[2].

[1]European Commission, Joint Research Centre, Geel, Belgium

[2]Oak Ridge National Laboratory, US. Department of Energy, Oak Ridge, TN, USA

Abstract

This report describes the re-certification of the IRMM-183-187 series of uranium nitrate solution reference materials with isotope amount fractions $n(^{235}U)/n(U)$ ranging from 0.3 % - 4.5 %, 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 Joint Research Centre's Unit G.2 in Geel/Belgium, the International Atomic Energy Agency (IAEA), in Seibersdorf Austria, the Oak Ridge National Laboratory (ORNL), in Oak Ridge, USA and the Commissariat à l'Energie Atomique et aux Energies Alternatives (CEA), in GIF SUR YVETTE CEDEX, France.

The IRMM-183-187 series of reference materials were originally produced and certified in the 1980s to 1990s. The so-called "major" isotope ratios $n(^{235}U)/n(^{238}U)$ were certified using gas source mass spectrometry (GSMS) and the so-called "minor" isotope ratios $n(^{234}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$ using thermal ionization mass spectrometry (TIMS). The minor ratios were re-certified in 2005 [4], but since several years customers like the IAEA have reported discrepancies of their measurement results from the certified values. Therefore a re-certification project was started by JRC-G2 in 2019, which included a complete re-characterization of the IRMM-183-187 series. For the measurement of the major ratios the double spike method (TIMS/DS) [5] according to ASTM C1871 [6] and for the minor ratios the modified total evaporation method (TIMS/MTE) [7,8] according to ASTM C1832 [9] were used. New verification measurements were performed using TIMS/MTE by ORNL and CEA, and historical data for TIMS/MTE measurements on three instruments from 2012-2021 were obtained from the IAEA. Based on the good agreement of the verification measurements and historical data with the new measurement results obtained during the re-characterization at JRC-G2, the isotope ratios of the IRMM-183-187 series were re-certified in line with ISO 17034 [1].

The re-certified IRMM-183-187 series of reference materials is recommended for the calibration of instruments and methods used for the measurement of the isotopic composition of uranium.

The following values were assigned to IRMM-183:

URANIUM NITRATE SOLUTION IRMM-183		
	Isotope amount ratio	
	Certified value 1) [mol/mol]	Uncertainty 2) [mol/mol]
$n(^{234}U)/n(^{238}U)$ $n(^{235}U)/n(^{238}U)$ $n(^{236}U)/n(^{238}U)$	0.000019814 0.00321826 0.000148492	0.000000014 0.00000052 0.000000030
	Isotope amount fraction	
	Certified value 3) [mol/mol]	Uncertainty 2) [mol/mol]
$n(^{234}U)/n(U)$ $n(^{235}U)/n(U)$ $n(^{236}U)/n(U)$ $n(^{238}U)/n(U)$	0.000019747 0.00320739 0.000147991 0.99662487	0.000000013 0.00000051 0.000000030 0.00000051
	Isotope mass fraction	
	Certified value 3) 4) [g/g]	Uncertainty 2) [g/g]
$m(^{234}U)/m(U)$ $m(^{235}U)/m(U)$ $m(^{236}U)/m(U)$ $m(^{238}U)/m(U)$	0.000019415 0.00316701 0.000146750 0.9966666682	0.000000013 0.00000051 0.000000030 0.00000051
	Molar mass	
	Certified value 3) 4) [g/mol]	Uncertainty 2) [g/mol]
$M(U)$	238.0407668	0.0000035

1) The certified values are traceable to the International System of units (SI) via the gravimetrically prepared reference material 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 *M. Wang, W.J. Huang, F.G. Kondev, G.Audi, S. Naimi, "The AME 2020 atomic mass evaluation", Chinese Physics C 45(3): 030003-1 – 030003-512 (2020):*
$M(^{234}U)$ = (234.0409503 ± 0.0000024) g/mol (k = 2)
$M(^{235}U)$ = (235.0439281 ± 0.0000024) g/mol (k = 2)
$M(^{236}U)$ = (236.0455661 ± 0.0000024) g/mol (k = 2)
$M(^{238}U)$ = (238.0507869 ± 0.0000032) g/mol (k = 2)

The following values were assigned to IRMM-184:

URANIUM NITRATE SOLUTION IRMM-184		
	Isotope amount ratio	
	Certified value [1)] [mol/mol]	Uncertainty [2)] [mol/mol]
$n(^{234}U)/n(^{238}U)$	0.000053196	0.000000016
$n(^{235}U)/n(^{238}U)$	0.0072631	0.0000011
$n(^{236}U)/n(^{238}U)$	0.00000012410	0.00000000096
	Isotope amount fraction	
	Certified value [3)] [mol/mol]	Uncertainty [2)] [mol/mol]
$n(^{234}U)/n(U)$	0.000052810	0.000000015
$n(^{235}U)/n(U)$	0.0072103	0.0000011
$n(^{236}U)/n(U)$	0.00000012320	0.00000000095
$n(^{238}U)/n(U)$	0.9927368	0.0000011
	Isotope mass fraction	
	Certified value [3) 4)] [g/g]	Uncertainty [2)] [g/g]
$m(^{234}U)/m(U)$	0.000051925	0.000000015
$m(^{235}U)/m(U)$	0.0071199	0.0000011
$m(^{236}U)/m(U)$	0.00000012217	0.00000000095
$m(^{238}U)/m(U)$	0.9928281	0.0000011
	Molar mass	
	Certified value [3) 4)] [g/mol]	Uncertainty [2)] [g/mol]
$M(U)$	238.0288945	0.0000047

1) The certified values are traceable to the International System of units (SI) via the gravimetrically prepared reference material 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 *M. Wang, W.J. Huang, F.G. Kondev, G.Audi, S. Naimi, "The AME 2020 atomic mass evaluation", Chinese Physics C 45(3): 030003-1 – 030003-512 (2020):*
$M(^{234}U)$ = (234.0409503 ± 0.0000024) g/mol (k = 2)
$M(^{235}U)$ = (235.0439281 ± 0.0000024) g/mol (k = 2)
$M(^{236}U)$ = (236.0455661 ± 0.0000024) g/mol (k = 2)
$M(^{238}U)$ = (238.0507869 ± 0.0000032) g/mol (k = 2)

The following values were assigned to IRMM-185:

URANIUM NITRATE SOLUTION IRMM-185		
	Isotope amount ratio	
	Certified value 1) [mol/mol]	Uncertainty 2) [mol/mol]
$n(^{234}U)/n(^{238}U)$	0.000179659	0.000000039
$n(^{235}U)/n(^{238}U)$	0.0200659	0.0000032
$n(^{236}U)/n(^{238}U)$	0.000002907	0.000000012
	Isotope amount fraction	
	Certified value 3) [mol/mol]	Uncertainty 2) [mol/mol]
$n(^{234}U)/n(U)$	0.000176093	0.000000038
$n(^{235}U)/n(U)$	0.0196677	0.0000030
$n(^{236}U)/n(U)$	0.000002849	0.000000012
$n(^{238}U)/n(U)$	0.9801534	0.0000030
	Isotope mass fraction	
	Certified value 3) 4) [g/g]	Uncertainty 2) [g/g]
$m(^{234}U)/m(U)$	0.000173171	0.000000037
$m(^{235}U)/m(U)$	0.0194242	0.0000030
$m(^{236}U)/m(U)$	0.000002826	0.000000011
$m(^{238}U)/m(U)$	0.9803998	0.0000030
	Molar mass	
	Certified value 3) 4) [g/mol]	Uncertainty 2) [g/mol]
$M(U)$	237.9909371	0.0000097

1) The certified values are traceable to the International System of units (SI) via the gravimetrically prepared reference material 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 *M. Wang, W.J. Huang, F.G. Kondev, G.Audi, S. Naimi, "The AME 2020 atomic mass evaluation", Chinese Physics C 45(3): 030003-1 – 030003-512 (2020):*
$M(^{234}U) = (234.0409503 \pm 0.0000024)$ g/mol (k = 2)
$M(^{235}U) = (235.0439281 \pm 0.0000024)$ g/mol (k = 2)
$M(^{236}U) = (236.0455661 \pm 0.0000024)$ g/mol (k = 2)
$M(^{238}U) = (238.0507869 \pm 0.0000032)$ g/mol (k = 2)

The following values were assigned to IRMM-186:

URANIUM NITRATE SOLUTION IRMM-186		
	Isotope amount ratio	
	Certified value [1)] [mol/mol]	Uncertainty [2)] [mol/mol]
$n(^{234}U)/n(^{238}U)$	0.000293966	0.000000064
$n(^{235}U)/n(^{238}U)$	0.0307894	0.0000048
$n(^{236}U)/n(^{238}U)$	0.000033388	0.000000034
	Isotope amount fraction	
	Certified value [3)] [mol/mol]	Uncertainty [2)] [mol/mol]
$n(^{234}U)/n(U)$	0.000285094	0.000000062
$n(^{235}U)/n(U)$	0.0298602	0.0000046
$n(^{236}U)/n(U)$	0.000032380	0.000000033
$n(^{238}U)/n(U)$	0.9698223	0.0000046
	Isotope mass fraction	
	Certified value [3) 4)] [g/g]	Uncertainty [2)] [g/g]
$m(^{234}U)/m(U)$	0.000280399	0.000000061
$m(^{235}U)/m(U)$	0.0294943	0.0000045
$m(^{236}U)/m(U)$	0.000032120	0.000000033
$m(^{238}U)/m(U)$	0.9701932	0.0000045
	Molar mass	
	Certified value [3) 4)] [g/mol]	Uncertainty [2)] [g/mol]
$M(U)$	237.959793	0.000014

1) The certified values are traceable to the International System of units (SI) via the gravimetrically prepared reference material 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 *M. Wang, W.J. Huang, F.G. Kondev, G.Audi, S. Naimi, "The AME 2020 atomic mass evaluation", Chinese Physics C 45(3): 030003-1 – 030003-512 (2020):*
$M(^{234}U)$ = (234.0409503 ± 0.0000024) g/mol (k = 2)
$M(^{235}U)$ = (235.0439281 ± 0.0000024) g/mol (k = 2)
$M(^{236}U)$ = (236.0455661 ± 0.0000024) g/mol (k = 2)
$M(^{238}U)$ = (238.0507869 ± 0.0000032) g/mol (k = 2)

The following values were assigned to IRMM-187:

URANIUM NITRATE SOLUTION IRMM-187		
	Isotope amount ratio	
	Certified value [1)] [mol/mol]	Uncertainty [2)] [mol/mol]
n(^{234}U)/*n*(^{238}U) *n*(^{235}U)/*n*(^{238}U) *n*(^{236}U)/*n*(^{238}U)	0.000387298 0.0473430 0.000072049	0.000000091 0.0000075 0.000000038
	Isotope amount fraction	
	Certified value [3)] [mol/mol]	Uncertainty [2)] [mol/mol]
n(^{234}U)/*n*(U) *n*(^{235}U)/*n*(U) *n*(^{236}U)/*n*(U) *n*(^{238}U)/*n*(U)	0.000369628 0.0451831 0.000068762 0.9543785	0.000000087 0.0000068 0.000000036 0.0000068
	Isotope mass fraction	
	Certified value [3) 4)] [g/g]	Uncertainty [2)] [g/g]
m(^{234}U)/*m*(U) *m*(^{235}U)/*m*(U) *m*(^{236}U)/*m*(U) *m*(^{238}U)/*m*(U)	0.000363612 0.0446382 0.000068222 0.9549300	0.000000086 0.0000067 0.000000036 0.0000067
	Molar mass	
	Certified value [3) 4)] [g/mol]	Uncertainty [2)] [g/mol]
M(U)	237.913308	0.000021

1) The certified values are traceable to the International System of units (SI) via the gravimetrically prepared reference material 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 *M. Wang, W.J. Huang, F.G. Kondev, G.Audi, S. Naimi, "The AME 2020 atomic mass evaluation", Chinese Physics C 45(3): 030003-1 – 030003-512 (2020):*
$M(^{234}U)$ = (234.0409503 ± 0.0000024) g/mol (k = 2)
$M(^{235}U)$ = (235.0439281 ± 0.0000024) g/mol (k = 2)
$M(^{236}U)$ = (236.0455661 ± 0.0000024) g/mol (k = 2)
$M(^{238}U)$ = (238.0507869 ± 0.0000032) g/mol (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 (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 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.

2 The Preparation of the IRMM-183-187 Series

The certification of the isotope ratios for the IRMM-183-187 series in the 1980s to 1990s was performed in several procedural steps. The IRMM-183-187 series of certified uranium nitrate solutions was prepared by dissolution from the original EC-171 series of uranium oxides (U_3O_8), consisting of the materials called EC-171/031, /071, /194, /295, /446. The IRMM-183-187 series solutions were also originally known as EC 183-187 and contain about 1 g of uranium in about 5 ml of 1 mol / l nitric acid solution per unit. Small fractions from the oxide series EC-171 were also fluorinated into UF_6 gas, and then calibrated for the $n(^{235}U)/n(^{238}U)$ ratio by GSMS (Gas Source Mass Spectrometry) against a set of 10 UF_6 internal standards prepared by fluorination from gravimetrically prepared uranium oxide mixtures (see sample preparation scheme in Annex 1). The fluorinated fractions of the EC-171 series were also used later as calibrants for the certification of the IRMM-019 to IRMM-029 series of UF_6 reference materials. The IRMM-019 to IRMM-029 series has already been re-certified in 2014 [10] and 2018 [11]. The uranium nitrate solutions prepared from it, the IRMM-2019 to IRMM-2029 series, was certified in 2018 [11].

Since the IRMM-183-187 series of uranium solutions was prepared by dissolution of the EC-171 oxides, the $n(^{235}U)/n(^{238}U)$ ratios of the IRMM-183-187 series were certified with the same values as obtained from the UF_6 measurements of the fluorinated fractions. The minor ratios of the IRMM-183-187 series were previously certified by TIMS in the 1980s to 1990s, but at that time the uncertainties were much larger compared to uncertainties achievable by modern TIMS instruments and methods.

The minor ratios $n(^{234}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$ were re-certified in 2004 [4] using the advantages in terms of precision provided by the new TRITON TIMS instrument from Thermofisher at JRC-G2. But the new results for the minor ratios were still normalized to the major ratios $n(^{235}U)/n(^{238}U)$ obtained by UF_6 GSMS in the 1990s. However, some of the uncertainties for the new minor ratios were underestimated in 2004 by stating measurement precisions instead of uncertainties as required by the GUM [3]. The uncertainties for the minor ratios were updated in 2019 following the recommendations from the ASTM C1832 standard for TIMS/MTE (Modified Total Evaporation) measurements, in particular by adding a relative uncertainty contribution of 0.4 % (k=2) for the use of a secondary electron multiplier for measuring ^{236}U [8, 9].

Nevertheless, not only for the minor but also for the major ratios, significant differences between new measurement results from the certified values were observed, not only at the IAEA but also at JRC-G2.

Therefore, in 2019 a new project was inaugurated at JRC-G2 for a complete re-characterization and re-certification of the IRMM-183-187 series. The original plan was to use the TIMS/MTE method for the re-characterization of all ratios, and in addition the high-precision double spike method TIMS/DS for the major ratio of only IRMM-184. The applicable range for the TIMS/DS method was later-on extended towards samples with $n(^{236}U)/n(^{238}U)$ ratios of up to 5×10^{-4} without an increase of the relative uncertainty for the major ratio $n(^{235}U)/n(^{238}U)$ [6]. This allowed the re-characterization of the major ratios using the TIMS/DS method for the entire IRMM-183-187 series.

3 Characterization

The characterization of the major ratios $n(^{235}U)/n(^{238}U)$ was performed at JRC-G2 using the double spike method (TIMS/DS) [5, 6] and for the minor ratios $n(^{234}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$ using the modified total evaporation method (TIMS/MTE) [7, 8, 9]. The TIMS/MTE results for the minor ratios were normalized using the results of the major ratios from the double spike measurements. The gravimetrically prepared double spike reference material IRMM-3636a [5] was used as a calibrant for the mass fractionation correction for the TIMS/DS method, and also indirectly for the TIMS/MTE method. For quality control (QC) purposes IRMM-074 [12] was used for the major ratios. Concerning the minor ratios the gravimetrically prepared reference material IRMM-075/1 [13] was used for QC purposes. The certificates for IRMM-3636a, IRMM-074 and IRMM-075 series are shown in Annex 2.

The TIMS/MTE method was recently upgraded at JRC-G2 for the use of Faraday cup amplifiers with 10^{13} Ω resistors. This was done 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 $n(^{236}U)/n(^{238}U)$ [14]. The upgrade of the method required longer integration and waiting times for the measurement of the minor isotopes and an improved control of the instrument mass calibration. The details of the upgraded TIMS/MTE method and QC measurement results using IRMM-075 series are described in [14].

Following the ASTM standard C1832 for the TIMS/MTE method, for the minor ratios below 10^{-5} in case of the $n(^{236}U)/n(^{238}U)$ ratios of IRMM-184 and IRMM-185, a secondary electron multiplier (SEM) in combination with an energy filter was used for the measurement of ^{236}U. The inter-calibration of the SEM detector versus the Faraday multi-collector system was performed internally using the ^{234}U ion beam as described in C1832. Additional QC measurements were performed by comparative measurements of the $n(^{236}U)/n(^{238}U)$ ratio of IRMM-187 with a value of about 7.2×10^{-5}, using either the SEM detector or the Faraday cup amplifier with a 10^{13} Ω resistor.

The TIMS analyses for all 5 materials were performed on loading solutions taken from 3 ampoules with at least 4 replicate filament loadings. There was no case of disagreement between results for the 3 loading solutions. Therefore, for each of the 5 materials and the isotope ratios, the results for each of the loading solutions were combined by averaging over all individual measurements. In case of the major ratio $n(^{235}U)/n(^{238}U)$ of IRMM-184 historical data from double spike measurements performed between 2010 and 2021 using material from an additional 4th ampoule were included for the characterization. The final results of the characterization are shown in Table 1.

Table 1. Isotope ratios for the IRMM-183-187 series as the result of the re-characterization in 2021. The major ratios $n(^{235}U)/n(^{238}U)$ were determined using the TIMS/DS method according to ASTM C1871 and the minor ratios $n(^{234}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$ using the TIMS/MTE method according to ASTM C1832, subsequently normalized using the results of the major ratios from the double spike measurements. Uncertainties are given with a coverage factor of k = 2 and apply to the last two digits of the result.

Sample	$n(^{234}U)/n(^{238}U)$	$n(^{235}U)/n(^{238}U)$	$n(^{236}U)/n(^{238}U)$
IRMM-183	0.000019814 (14)	0.00321826 (52)	0.000148492 (30)
IRMM-184	0.000053196 (16)	0.0072631 (11)	0.00000012410 (96)
IRMM-185	0.000179659 (39)	0.0200659 (32)	0.000002907 (12)
IRMM-186	0.000293966 (64)	0.0307894 (48)	0.000033388 (34)
IRMM-187	0.000387298 (91)	0.0473430 (75)	0.000072049 (38)

The relative uncertainties for the major ratios $n(^{235}U)/n(^{238}U)$ are at a level of 0.016 % (k=2) and are strongly (>95%) dominated by the uncertainty of the certified $n(^{233}U)/n(^{236}U)$ ratio of the double spike IRMM-3636a. This is due to the very good relative precision (i.e. relative standard deviation) among replicate filament measurements, which is at a level of < 0.005 %. The uncertainty contribution from the $n(^{236}U)/n(^{238}U)$ ratio of the samples of IRMM-183-187 for the isobaric correction of the measured $n(^{233}U)/n(^{236}U)$ ratio was negligible, due to the low $n(^{236}U)/n(^{238}U)$ ratios (< 5×10^{-4} [6]) and their small uncertainties from the MTE measurements.

The minor ratios were determined using the TIMS/MTE method and subsequently normalized using the results of the major ratios from the double spike measurements. As explained in detail in [8] and the ASTM standard C1832 [9], the uncertainties for the minor ratios depend strongly on the order of magnitude of the ratios. Uncertainty budgets for the characterization of the minor ratios are presented in Table 2 and Table 3.

In the case of $n(^{234}U)/n(^{238}U)$, the uncertainties improve along the IRMM-183-187 series as expected. Except for IRMM-183, where the uncertainty of the IRMM-3636a double spike is dominating the uncertainty budget over the measurement repeatability.

In case of $n(^{236}U)/n(^{238}U)$, the observation is similar among Faraday cup measurements, but the $n(^{236}U)/n(^{238}U)$ ratios for IRMM-184 and IRMM-185 are below 10^{-5} and were therefore measured using the SEM in ion counting mode, with an additional relative expanded uncertainty of 0. 4 % included for the inter-calibration of the SEM versus the Faraday multi-collector system (coverage factor k=2). Furthermore, an uncertainty contribution for the background correction at mass 236 was included, based on measurements of natural uranium samples [15] with $n(^{236}U)/n(^{238}U)$ ratios at the level of $< 2 \times 10^{-10}$. The background at mass 236 is caused by un-identified interferences from the ionization filament.

Table 2. Uncertainty budgets for the "minor" ratios $n(^{234}U)/n(^{238}U)$ for the IRMM-183-187 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-183	**IRMM-184**	**IRMM-185**	**IRMM-186**	**IRMM-187**
Ratio $n(^{234}U)/n(^{238}U)$	0.000019814	0.000053196	0.000179659	0.000293966	0.000387298
Rel. Unc. (k=2), in %	0.068	0.029	0.022	0.022	0.024
Relative uncertainty contribution in % (*)					
Contribution from measurement of $n(^{234}U)/n(^{238}U)$	90.1	48.2	5.1	6.3	20.0
IRMM-3636a certified $n(^{233}U)/n(^{236}U)$ ratio	9.4	50.9	93.7	93.4	79
Measurement of $n(^{235}U)/n(^{238}U)$ within TIMS/DS	0.4	0.7	1.0	0.1	0.8
Peak tailing for Faraday detection	0.0	0.2	0.2	0.2	0.2

Table 3. Uncertainty budgets for the "minor" ratios $n(^{236}U)/n(^{238}U)$ for the IRMM-183-187 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-183	**IRMM-184**	**IRMM-185**	**IRMM-186**	**IRMM-187**
Ratio $n(^{236}U)/n(^{238}U)$	0.000148492	1.241×10^{-7}	0.000002907	0.000033388	0.000072049
Rel. Unc. (k=2), in %	0.020	0.77	0.41	0.10	0.052
Relative Uncertainty contribution in %(*)					
Contribution from measurement of $n(^{236}U)/n(^{238}U)$	51.3	3.7	1.8	49.3	49.7
IRMM-3636a certified $n(^{233}U)/n(^{236}U)$ ratio	26.9	0.0	0.1	1.0	4.0
Measurement of $n(^{235}U)/n(^{238}U)$ within TIMS/DS	1.3	0.0	0.0	0.0	0.0

Peak tailing for Faraday detection	20.6	N/A	N/A	49.7	46.3
SEM calibration	N/A	26.7	97.7	N/A	N/A
Background ^{236}U	N/A	69.6	0.5	N/A	N/A

4 Quality Control Measurements at JRC-G2

In Figure 1 and Figure 2, QC charts from JRC-G2 are presented to confirm that the major ratios $n(^{235}U)/n(^{238}U)$ measured using the TIMS/DS method are stable over time periods of at least 10 years. The control limits are based on experience at JRC-G2 and the measurement performance is documented in [6]. This underpins the 10 years validity of the certified isotope ratios of the IRMM-183-187 series obtained using this method. In particular, for the gravimetrically prepared IRMM-074 series (basic mixture of ^{235}U and ^{238}U oxides, [12]), the QC chart shows the level of trueness for TIMS/DS measurements using the IRMM-3636a double spike reference material. The same material is also used for the certification of the IRMM-183-187 series.

The QC chart for IRMM-184 in Figure 2 shows a 12-year average relative difference of the measured $n(^{235}U)/n(^{238}U)$ ratios from a literature reference value published in 2010 [16] of only -0.000024 % and a relative standard deviation of the ratios of only s = 0.0041 %. This reference value was established in 2010 by an inter-comparison of double spike measurements among several expert laboratories and has been used by many laboratories since, instead of the previous certified value. The new certified value for the $n(^{235}U)/n(^{238}U)$ ratio of IRMM-184 is the average all the double spike measurements performed over 12 years at JRC-G2. It is not equal but very close to the literature reference value.

In Figure 3, a QC chart for $n(^{236}U)/n(^{238}U)$ ratio measurements within a period of more than 16 years for the gravimetrically prepared IRMM-075/1 [13] with a certified value of $n(^{236}U)/n(^{238}U)$ = 0.000104433(37) (k=2) using TIMS/MTE by JRC-G2l is shown. The precision is significantly better, and the uncertainties are lower, when 10^{13} Ω amplifiers are used, as explained in detail in [14]. The performance of $n(^{234}U)/n(^{238}U)$ ratio measurements was monitored in a similar manner using IRMM-075/1, and is explained in more detail in [17].

Figure 1. 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 [12]) using TIMS/DS by JRC-G2. The uncertainties are given with coverage factor k=2.

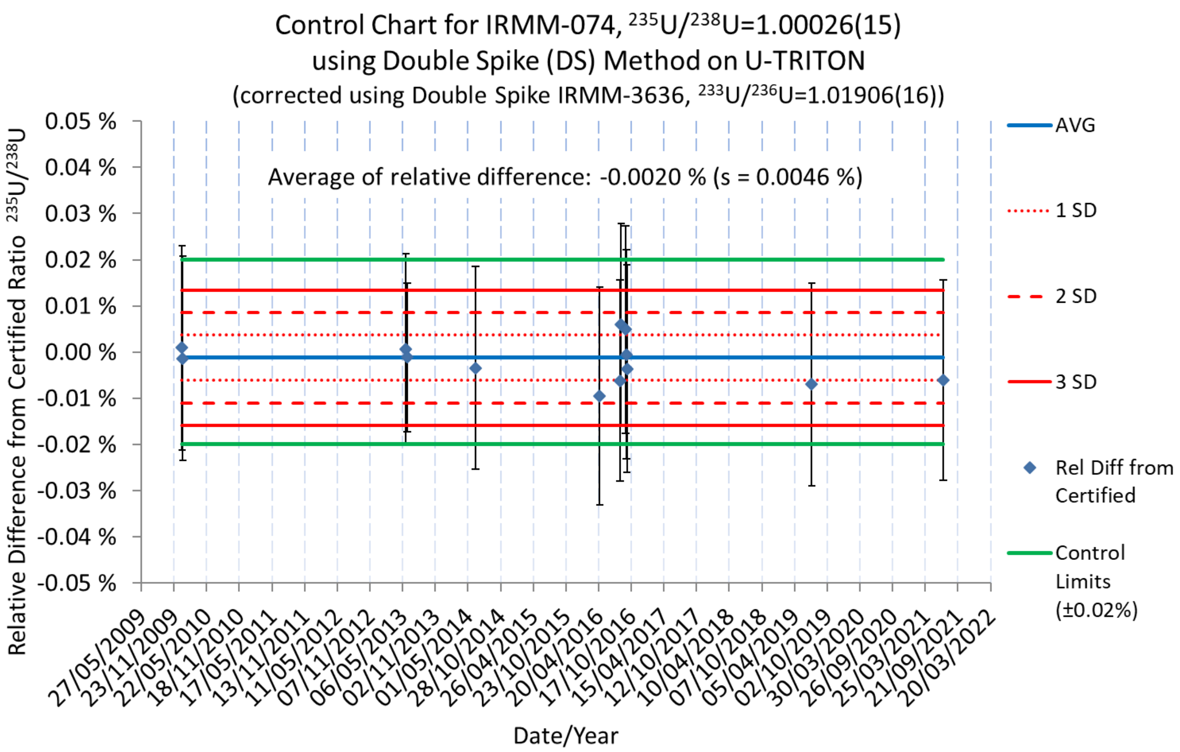


Figure 2. QC chart for $n(^{235}U)/n(^{238}U)$ ratio measurements of IRMM-184 using TIMS/DS by JRC-G2. The uncertainties are given with coverage factor k=2.

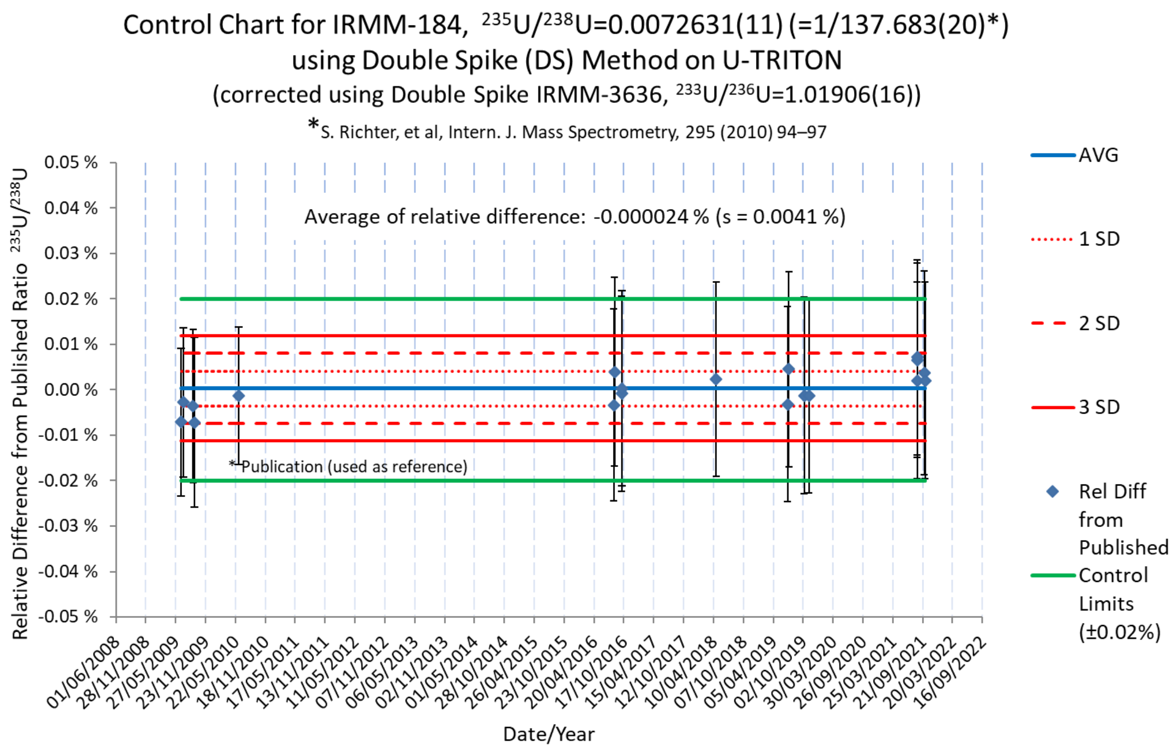
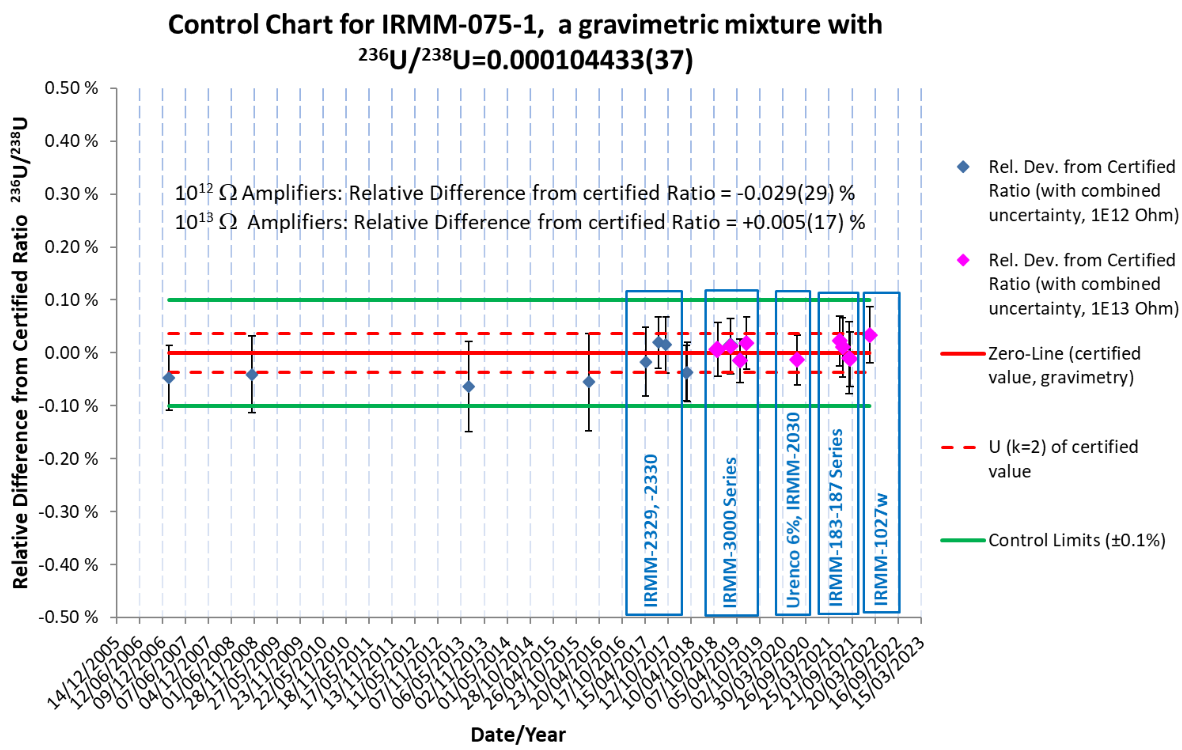


Figure 3. QC measurements for the $n(^{236}U)/n(^{238}U)$ ratios for IRMM-075/1. The uncertainties are given with coverage factor k=2.



5 Verification Measurements by External Collaborators

Verification measurements for the isotopic composition for the entire IRMM-183-187 series were performed using the TIMS/MTE method by JRC-G2, ORNL and the-SGAS/NML. The results from JRC-G2 and ORNL are normalized using IRMM-074 [12] and the new IRMM-3050 [17], respectively. From the IAEA, historical TIMS/MTE data acquired on three TRITON TIMS instruments operated within the NML between 2012 and 2021 were received. They were all originally normalized using the previous certified $n(^{235}\text{U})/n(^{238}\text{U})$ ratio IRMM-184. This previous value only differs slightly 0.01 %) and insignificantly from the new proposed certified ratio. Therefore a re-normalization of the original verification data from the IAEA was not justified. Furthermore, from CEA-LANIE un-normalized historical data for all isotope ratios of IRMM-187 and the $n(^{234}\text{U})/n(^{238}\text{U})$ ratio of IRMM-184 measured using TIMS/MTE were collected. They have retroactively been normalized using the new certified $n(^{235}\text{U})/n(^{238}\text{U})$ ratio.

In Figure 4 to Figure 18 the results for the verification measurements are presented. Each data point represents the result of an automatic measurement sequence performed on one sample magazine with 21 filaments, measured on one instrument in a particular facility as indicated on the abscissa, with some sequences repeated in a similar manner for various reasons. The different amplifiers equipped with $10^{11}\ \Omega$, $10^{12}\ \Omega$ or $10^{13}\ \Omega$ resistors are indicated in the figures as “1E11”, “1E12” and “1E13”. The SEM detector is also used for measuring some of the $n(^{236}\text{U})/n(^{238}\text{U})$ ratios, at JRC-G2 and ORNL only for IRMM-184 and IRMM-185, and at the IAEA for the entire series.

Figure 4. Verification of $n(^{234}\text{U})/n(^{238}\text{U})$ ratio for IRMM-183 by the JRC-G2, IAEA/NML and ORNL. The dashed lines represent the relative expanded uncertainties of the certified ratios. All uncertainties are given with coverage factor k=2.

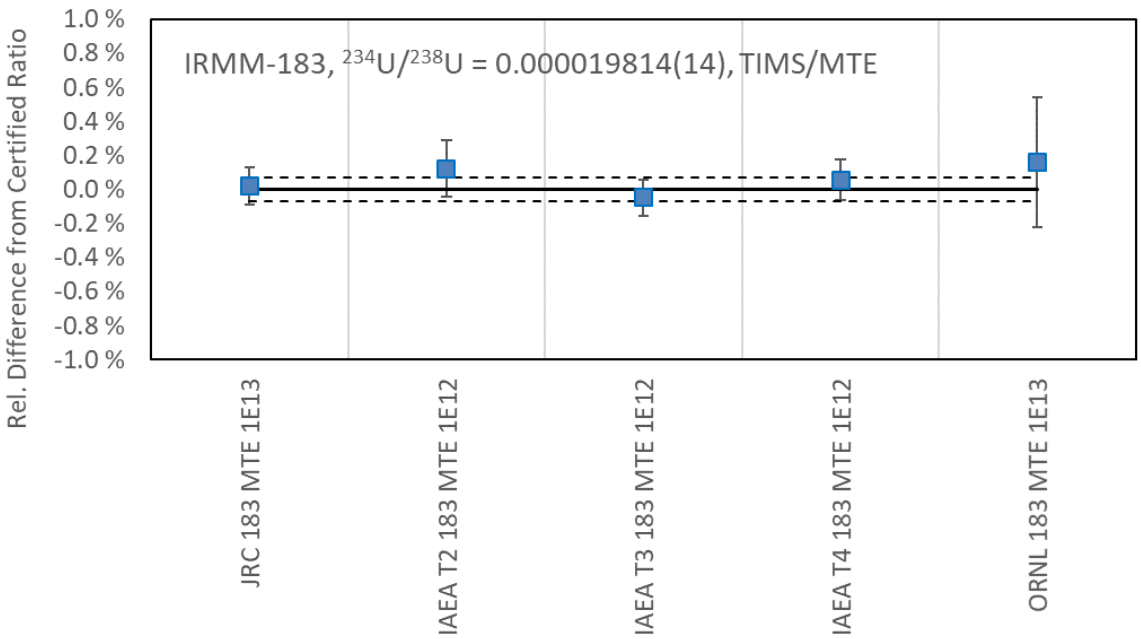


Figure 5. Verification of $n(^{234}U)/n(^{238}U)$ ratio for IRMM-184 by the JRC-G2, IAEA/NML, ORNL and CEA. The dashed lines represent the relative expanded uncertainties of the certified ratios. All uncertainties are given with coverage factor k=2.

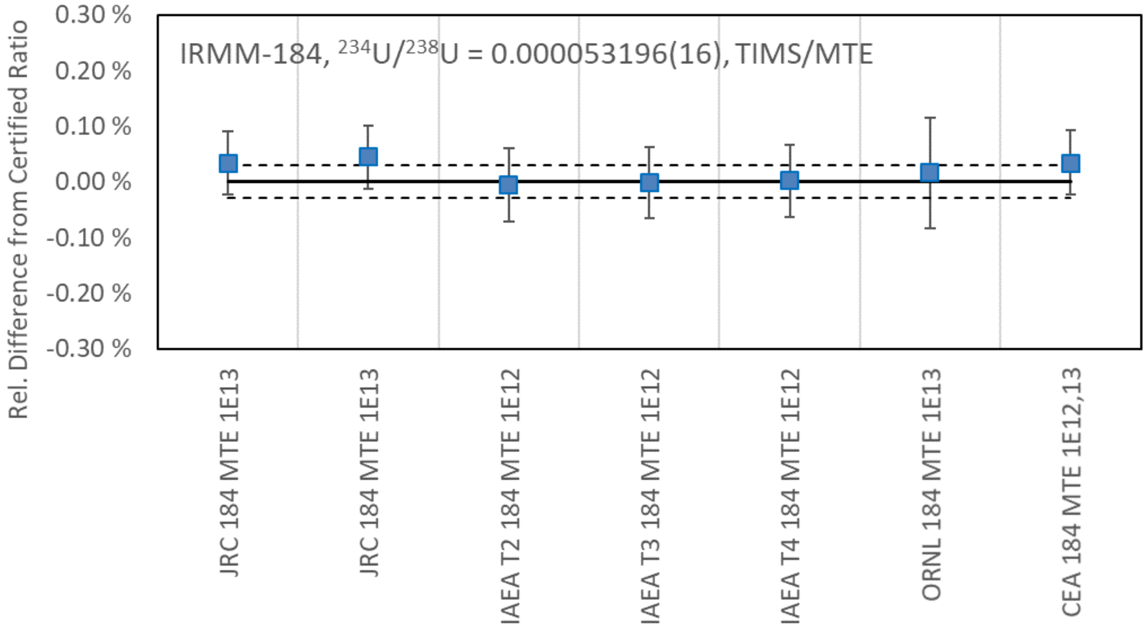


Figure 6. Verification of $n(^{234}U)/n(^{238}U)$ ratio for IRMM-185 by the JRC-G2, IAEA/NML and ORNL. The dashed lines represent the relative expanded uncertainties of the certified ratios. All uncertainties are given with coverage factor k=2.

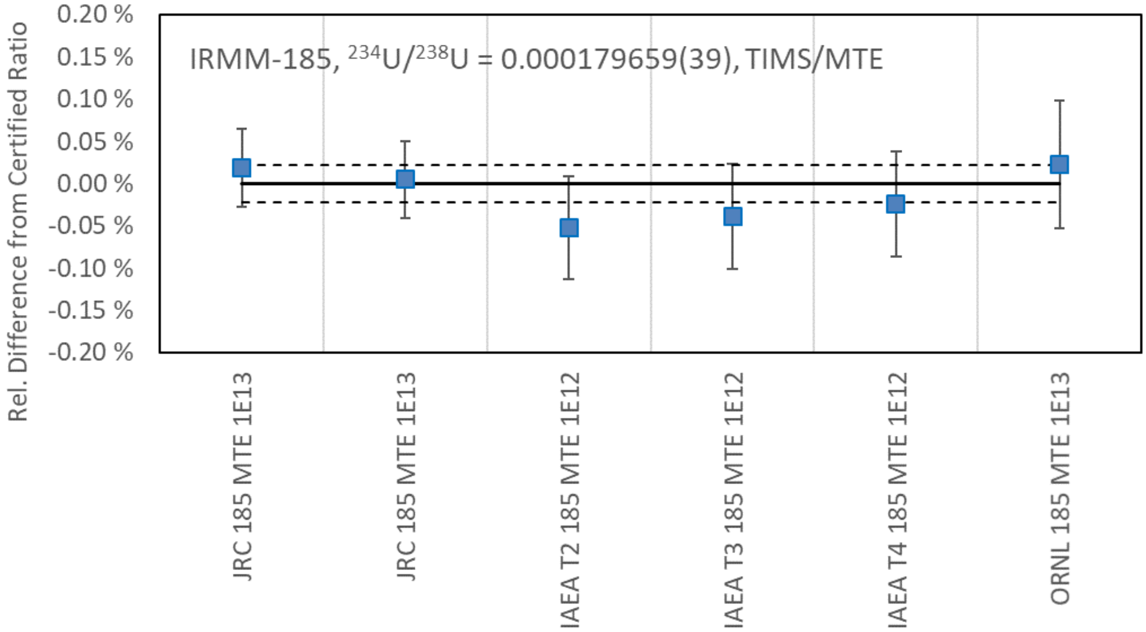


Figure 7. Verification of $n(^{234}U)/n(^{238}U)$ ratio for IRMM-186 by the JRC-G2, IAEA/NML and ORNL. The dashed lines represent the relative expanded uncertainties of the certified ratios. All uncertainties are given with coverage factor k=2.

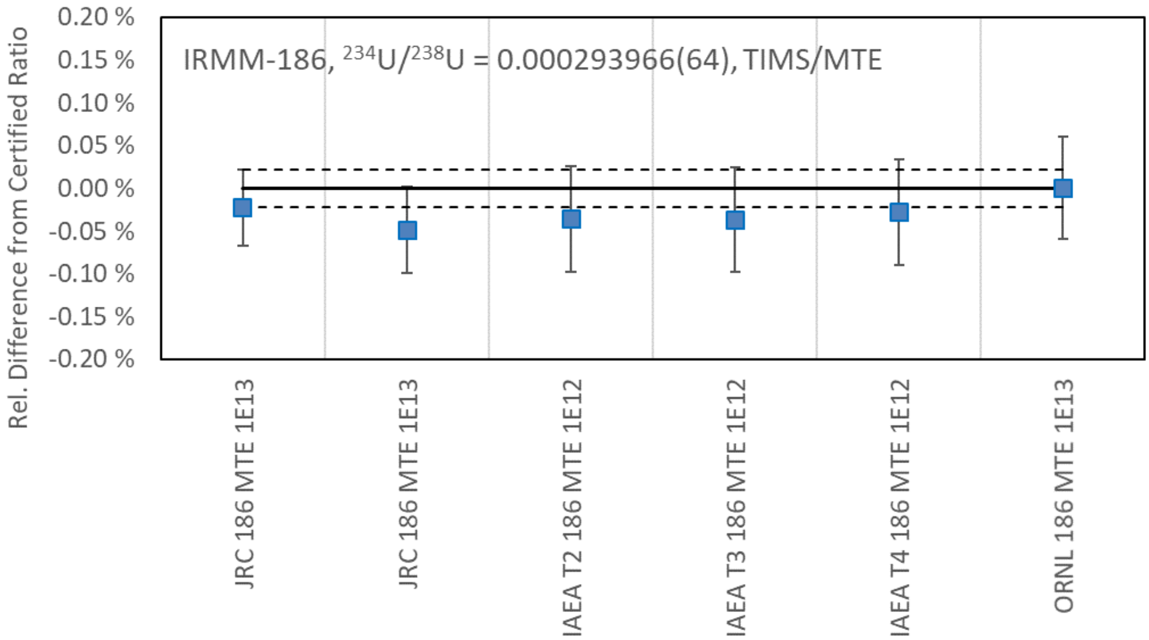


Figure 8. Verification of $n(^{234}U)/n(^{238}U)$ ratio for IRMM-187 by the JRC-G2, IAEA/NML, ORNL and CEA. The dashed lines represent the relative expanded uncertainties of the certified ratios. All uncertainties are given with coverage factor k=2.

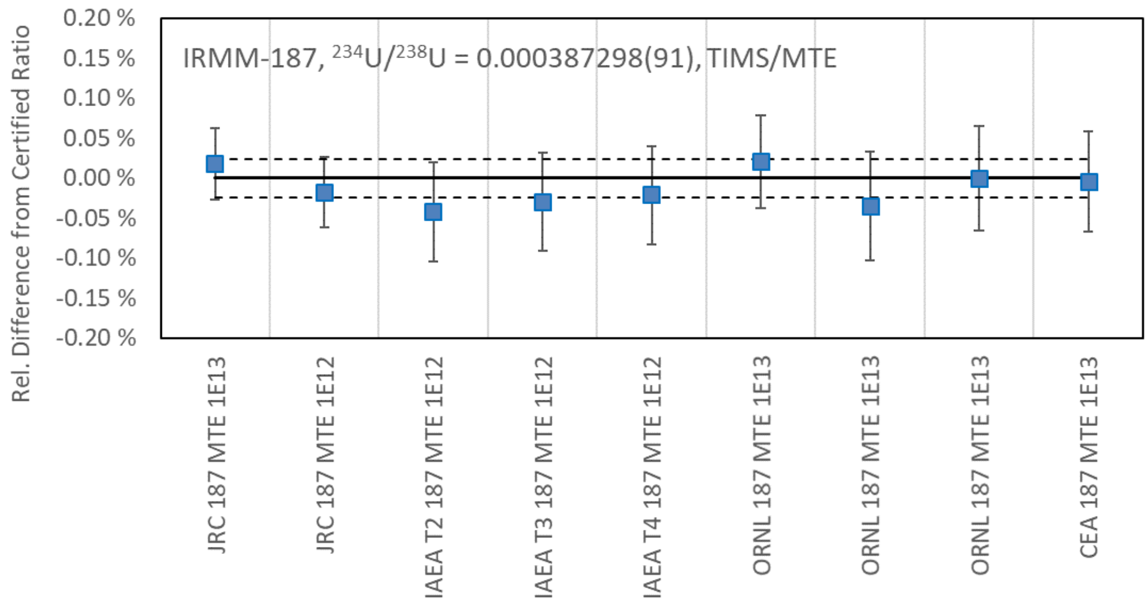


Figure 9. Verification of $n(^{235}U)/n(^{238}U)$ ratio for IRMM-183 by the JRC-G2, IAEA/NML and ORNL. The dashed lines represent the relative expanded uncertainties of the certified ratios. All uncertainties are given with coverage factor k=2.

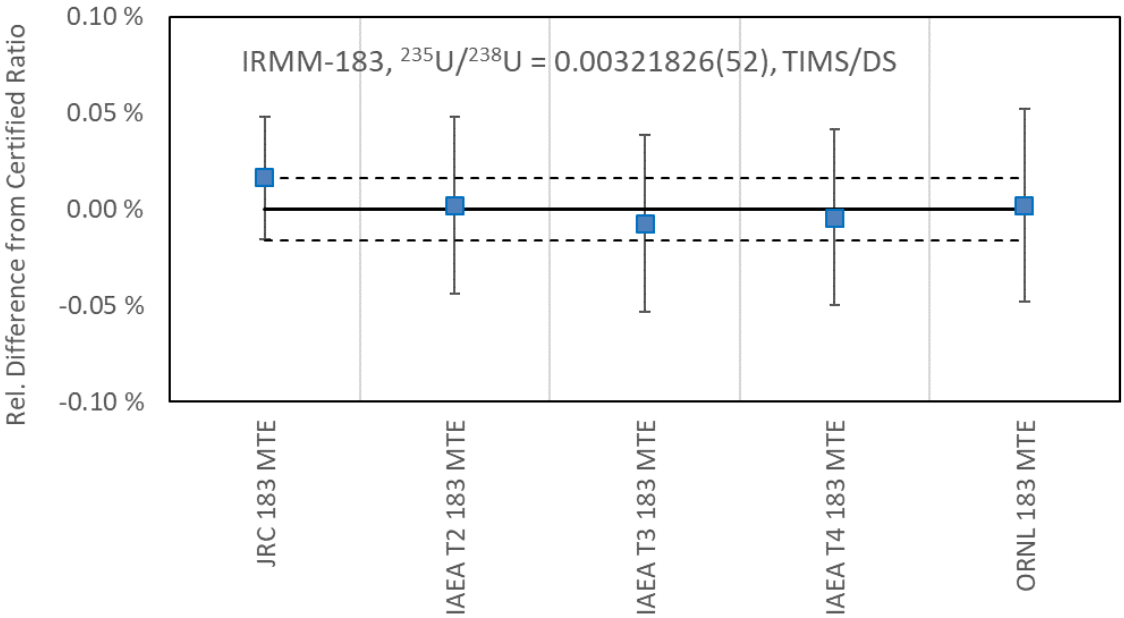


Figure 10. Verification of $n(^{235}U)/n(^{238}U)$ ratio for IRMM-184 by the JRC-G2, IAEA/NML and ORNL. The dashed lines represent the relative expanded uncertainties of the certified ratios. All uncertainties are given with coverage factor k=2.

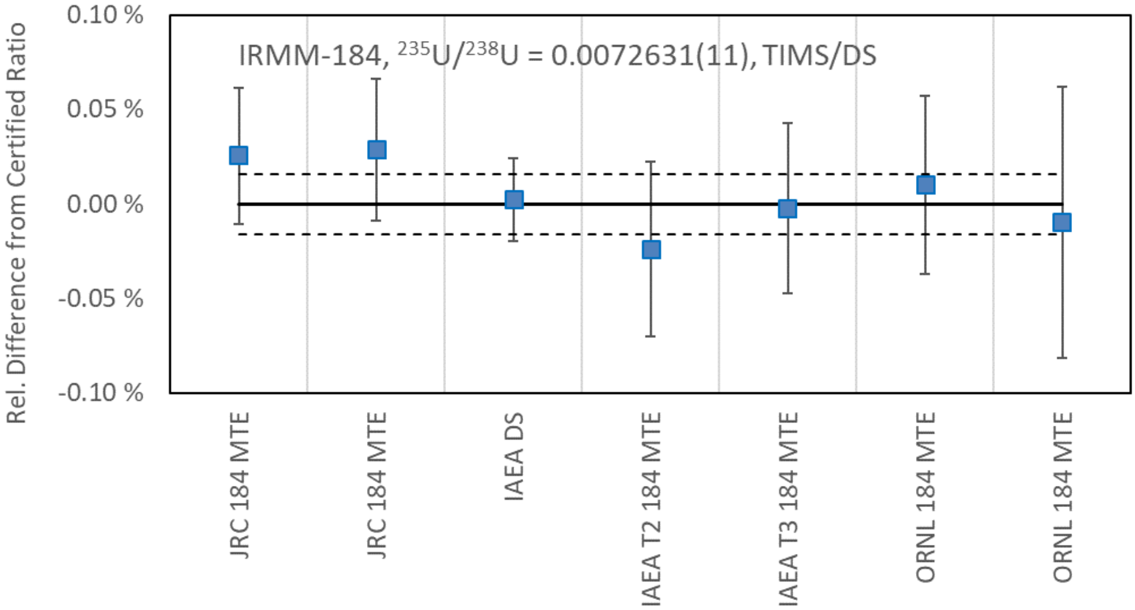


Figure 11. Verification of $n(^{235}U)/n(^{238}U)$ ratio for IRMM-185 by the JRC-G2, IAEA/NML and ORNL. The dashed lines represent the relative expanded uncertainties of the certified ratios. All uncertainties are given with coverage factor k=2.

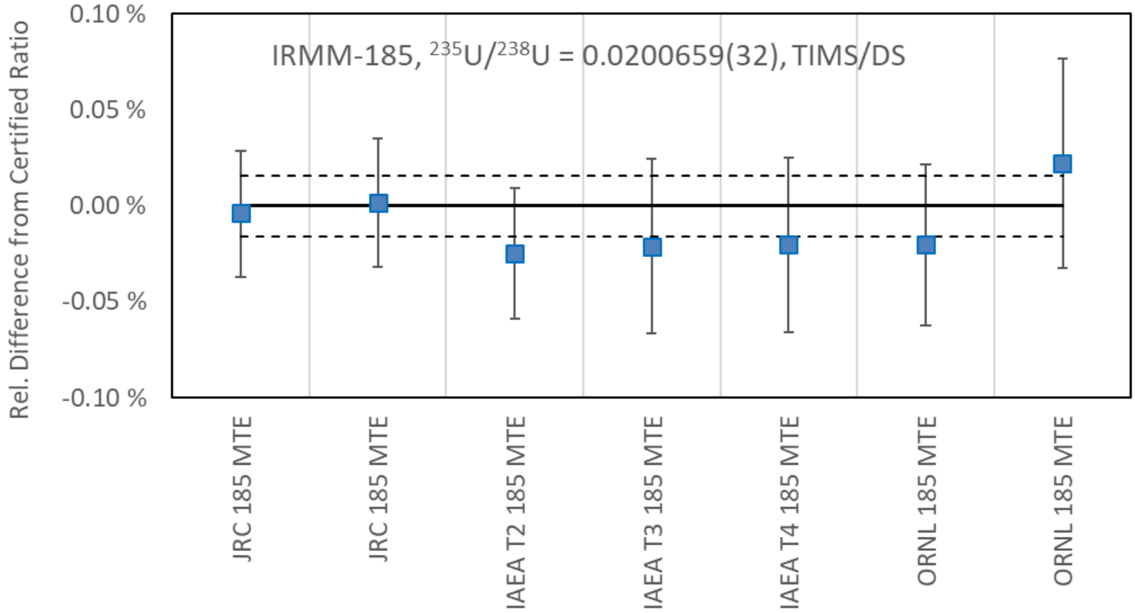


Figure 12. Verification of $n(^{235}U)/n(^{238}U)$ ratio for IRMM-186 by the JRC-G2, IAEA/NML and ORNL. The dashed lines represent the relative expanded uncertainties of the certified ratios. All uncertainties are given with coverage factor k=2.

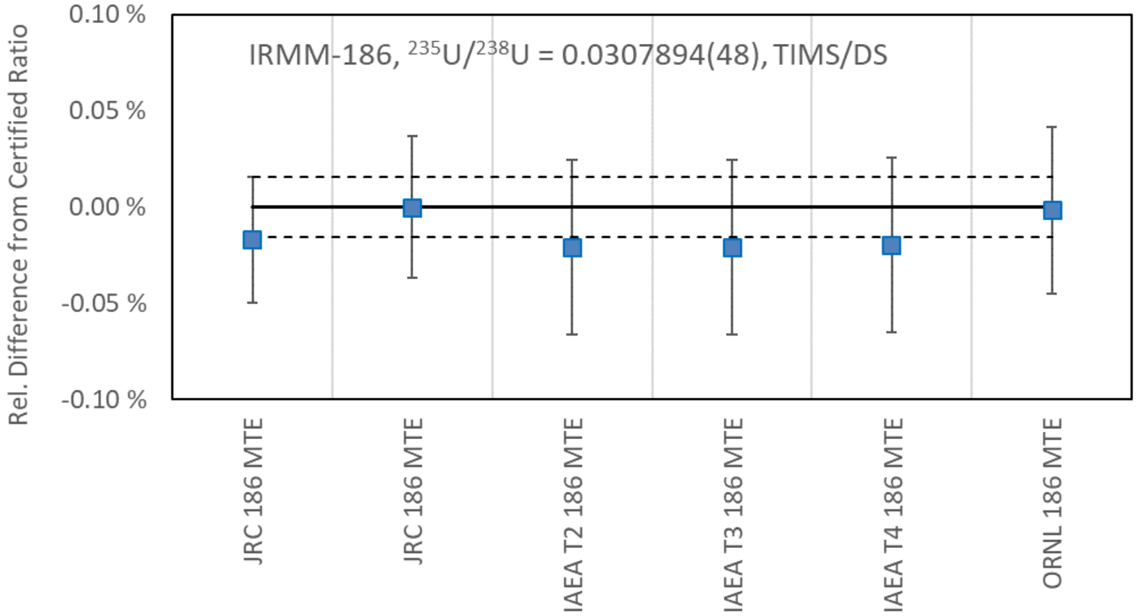


Figure 13. Verification of $n(^{235}U)/n(^{238}U)$ ratio for IRMM-187 by the JRC-G2, IAEA/NML, ORNL and CEA. The dashed lines represent the relative expanded uncertainties of the certified ratios. All uncertainties are given with coverage factor k=2.

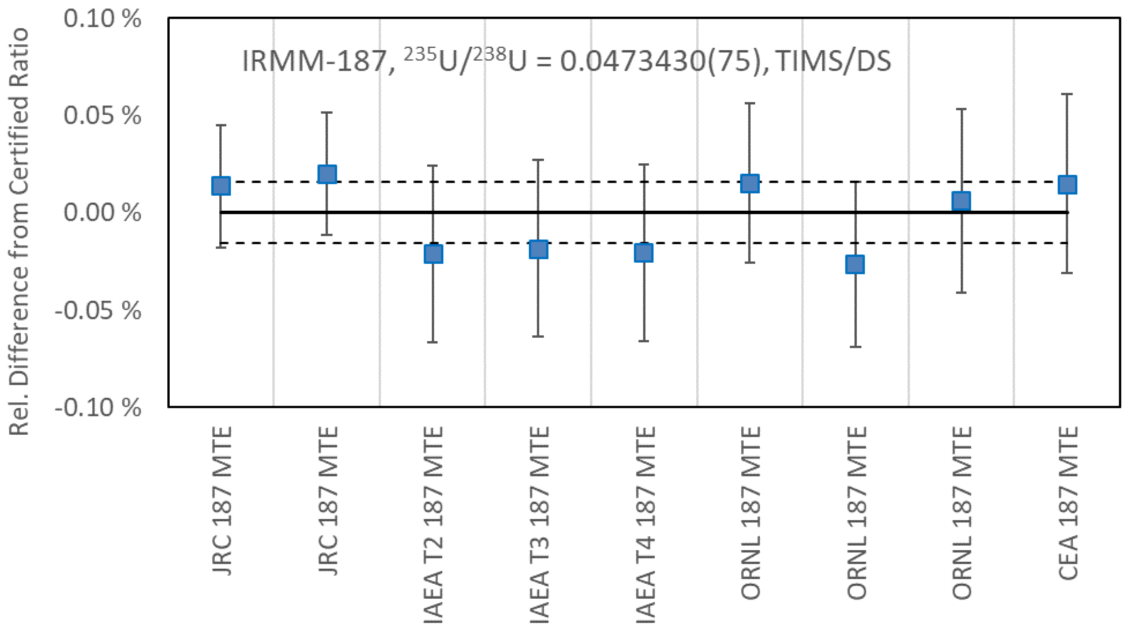


Figure 14. Verification of $n(^{236}U)/n(^{238}U)$ ratio for IRMM-183 by the JRC-G2, IAEA/NML and ORNL. The dashed lines represent the relative expanded uncertainties of the certified ratios. All uncertainties are given with coverage factor k=2.

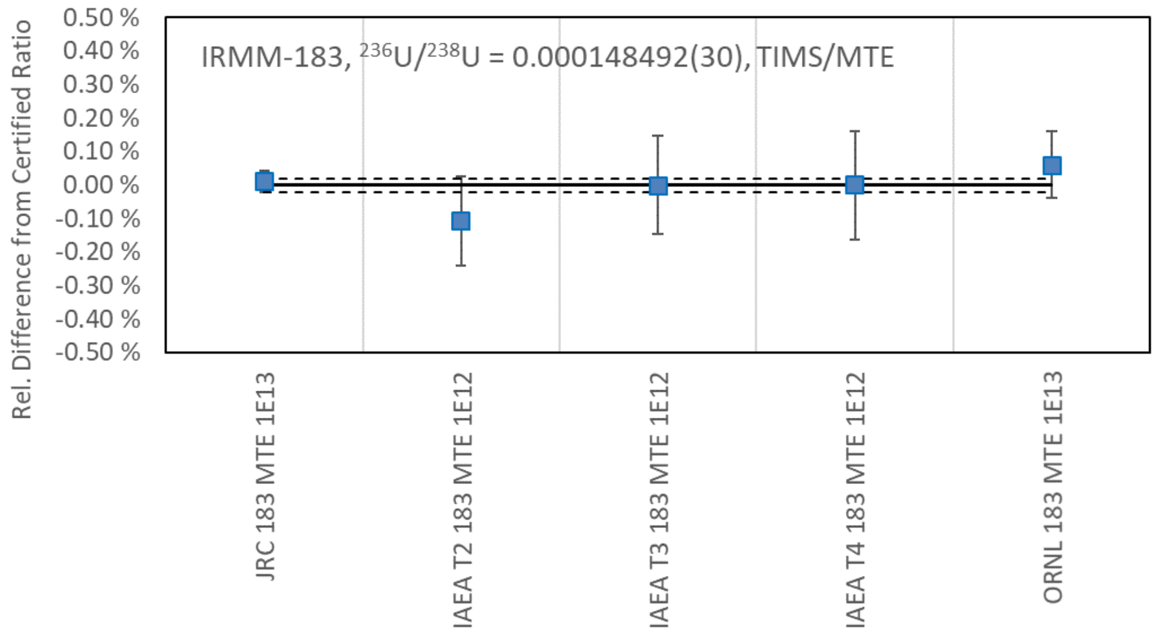


Figure 15. Verification of $n(^{236}U)/n(^{238}U)$ ratio for IRMM-184 by the JRC-G2, IAEA/NML and ORNL. The dashed lines represent the relative expanded uncertainties of the certified ratios. All uncertainties are given with coverage factor k=2.

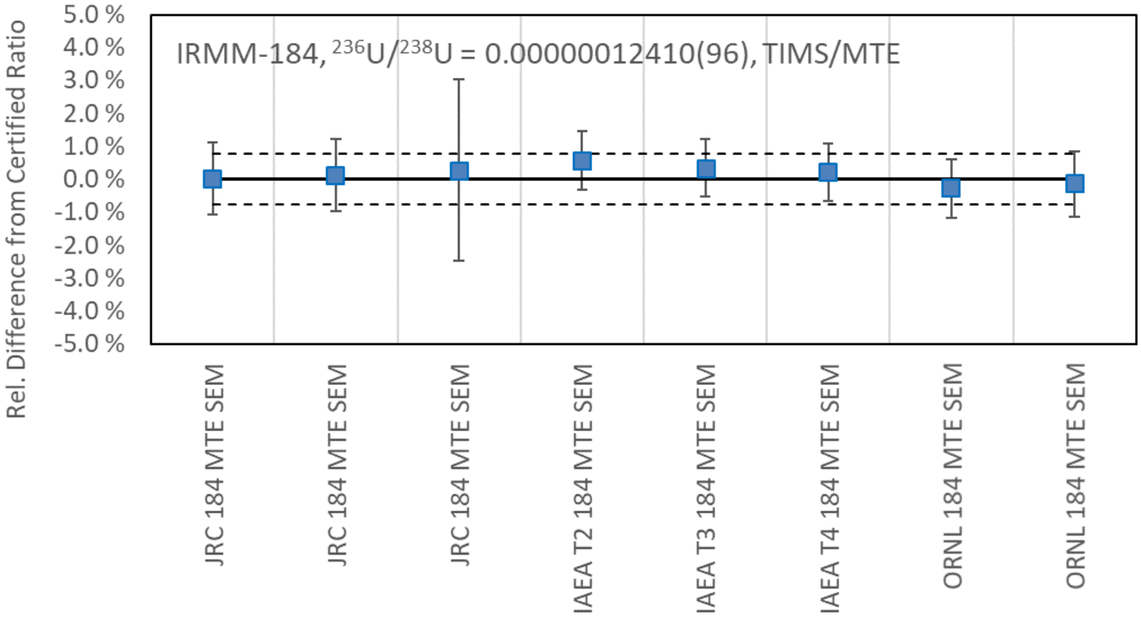


Figure 16. Verification of $n(^{236}U)/n(^{238}U)$ ratio for IRMM-185 by the JRC-G2, IAEA/NML and ORNL. The dashed lines represent the relative expanded uncertainties of the certified ratios. All uncertainties are given with coverage factor k=2.

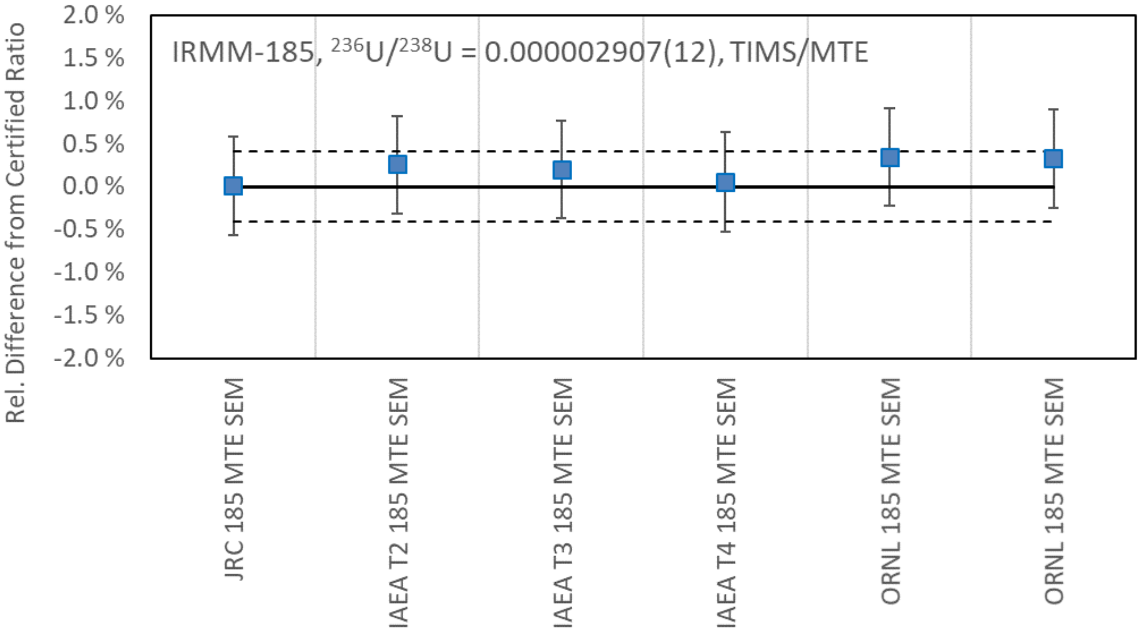


Figure 17. Verification of $n(^{236}U)/n(^{238}U)$ ratio for IRMM-186 by the JRC-G2, IAEA/NML and ORNL. The dashed lines represent the relative expanded uncertainties of the certified ratios. All uncertainties are given with coverage factor k=2.

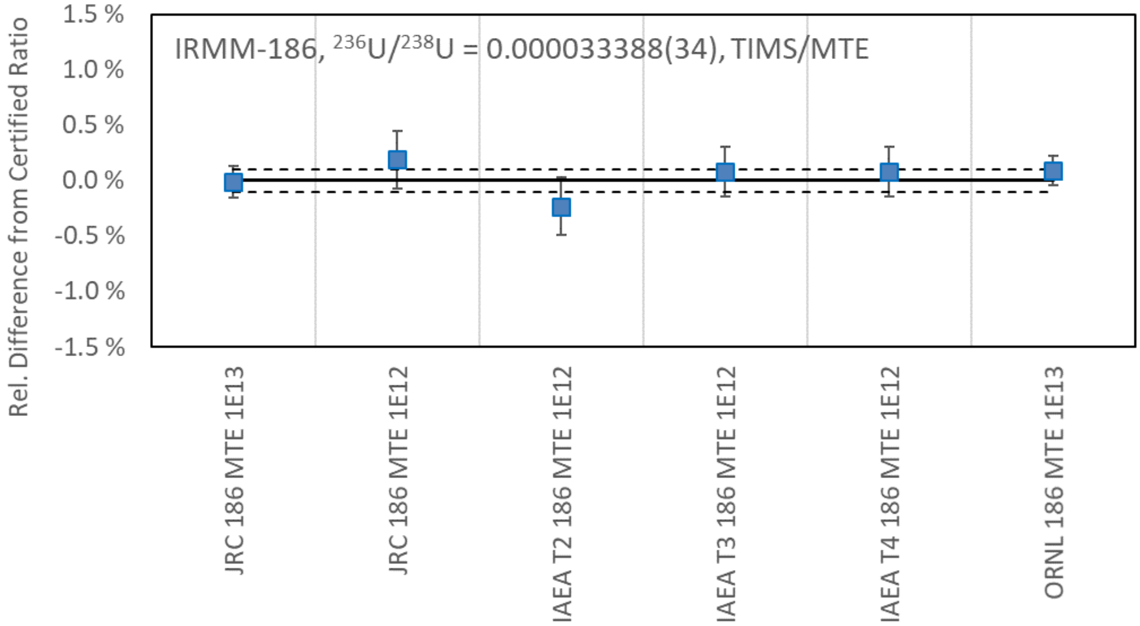
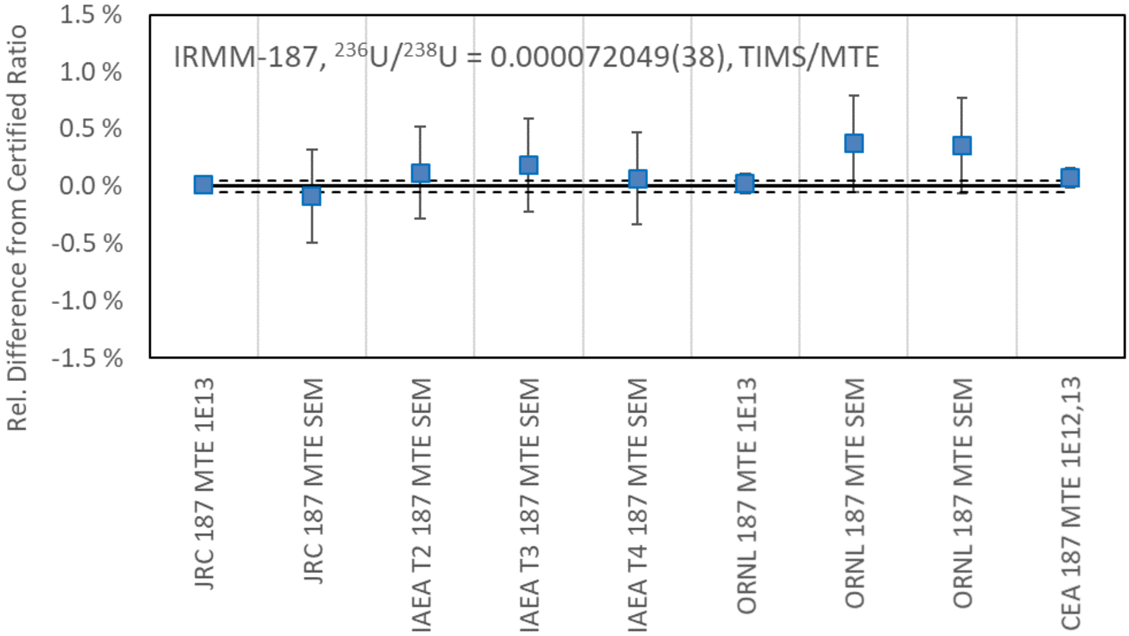


Figure 18. Verification of $n(^{236}U)/n(^{238}U)$ ratio for IRMM-187 by the JRC-G2, IAEA/NML, ORNL and CEA. The dashed lines represent the relative expanded uncertainties of the certified ratios. All uncertainties are given with coverage factor k=2.



The results for the relative differences 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 using the TIMS/MTE method by the JRC-G2, IAEA, ORNL and CEA demonstrate agreement with the certified values proposed by JRC-G2 within the stated uncertainties. The measurement uncertainties encountered for the reported TIMS/MTE measurements are in line with observations from the ASTM C1832 standard document [9]. In addition to the IRMM-183-187 series, also various different reference materials were measured as QC samples by the same laboratories and agree well with the respective certified values.

Remarkably, for the historical data for 3 TIMS instruments at the IAEA/NML, the major ratios $n(^{235}U)/n(^{238}U)$ tend to be slightly (but not significantly) below the new certified ratios obtained by the TIMS/DS spike method at JRC-G2, in particular for IRMM-185, IRMM-186 and IRMM-187 (see Figure 11, Figure 12 and Figure 13). This could be related to the normalization of the IAEA results using the previous certified $n(^{235}U)/n(^{238}U)$ ratio for IRMM-184, which is about 0.01% lower than the new certified $n(^{235}U)/n(^{238}U)$ ratio.

In case of $n(^{236}U)/n(^{238}U)$, the ratios of IRMM-184 and IRMM-185 are below 10^{-5} and were measured using the SEM in ion counting mode, with an additional relative uncertainty of 0.4 % (k=2) [8, 9] included for the inter-calibration of the SEM versus the Faraday multi-collector system. As shown in Figure 18, the validity of this inter-calibration was verified successfully at JRC-G2 and ORNL by measuring the $n(^{236}U)/n(^{238}U)$ ratio of about 7×10^{-5} for IRMM-187 on both the SEM and the Faraday cup multi-collector system. The relative standard deviation among all $n(^{236}U)/n(^{238}U)$ ratios obtained using the SEM is about 0.17 %, which is similar to the relative uncertainties (at a level of k = 1) of about 0.2 % for these ratios. This confirms that the 0.4 % uncertainty contribution (with k = 2) for the inter-calibration derived in [8] and recommended in the ASTM standard C1832 [9] is indeed realistic. The uncertainties for the $n(^{236}U)/n(^{238}U)$ ratios obtained using Faraday cups with 10^{13} Ω resistors as provided by JRC-G2, ORNL and CEA are at a level of 0.06 % and therefore much lower compared to those measured using the SEM, and their relative standard deviation is about 0.033 %, which is (also) similar to the relative uncertainties (k = 1) of about 0.029 % of these ratios. Therefore the $n(^{236}U)/n(^{238}U)$ratios obtained using Faraday cups with 10^{13} Ω resistors are used for the certification .

As a conclusion, the verification measurements by all external collaborators were successful. Therefore the results of the re-characterization measurements performed at JRC-G2 can be used for the re-certification of the IRMM-183-187 series.

6 Comparison of the new certified ratios with the previous ones

In Figure 19, Figure 20 and Figure 21 the new certified ratios of the IRMM-183-187 series (see also Table 1) are compared to the previous ones from 2019. The uncertainties of the relative differences between the new certified values versus the previous certified values already include the uncertainties for both of them. The previous certified $n(^{235}U)/n(^{238}U)$ ratios were based on UF_6 GSMS measurements performed in the 1980s and 1990s and re-measurements of the minor ratios $n(^{234}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$ in 2004, with the uncertainties of the $n(^{236}U)/n(^{238}U)$ ratios updated according to ASTM C1832 [9] in 2019.

As shown in Figure 19, for the $n(^{234}U)/n(^{238}U)$ ratio significant differences can be observed for the entire IRMM-183-187 series, in particular for IRMM-183 (0.3 %) but also for all other materials at a level of about 0.1 %. These differences can be explained by the change from the arithmetic peak tailing correction used in 2004 [4] towards the geometric (i.e. logarithmic) peak tailing correction applied for the more recent certification measurements using 10^{13} Ω amplifiers, as for the IRMM-3000 series [17] and the re-certification of the IRMM-183-187 series. A detailed explanation of the peak tailing correction is presented in Annex 9 of [17]. The arithmetic peak tailing correction usually causes an overestimation of the peak tailing contribution and therefore leads to too low values for the corrected $n(^{234}U)/n(^{238}U)$ ratios, whereas the logarithmic peak tailing correction also takes the curvature of the peak tail into consideration and is therefore more realistic.

The reason why the geometric (i.e. logarithmic) peak tailing correction was not used in 2004, at a time when only 10^{11} Ω amplifiers with much more noise (factor 5-10) compared to 10^{13} Ω amplifiers were available, is the fact, that the higher noise caused a more significant fraction of the background measurements to be negative on a statistical basis. This made the calculation of the geometric mean for the tailing correction impossible, because this was leading to a too large and unacceptable number of technical outlier measurement cycles during the measurements. With the availability of 10^{13} Ω amplifiers with much lower noise this problem has been resolved.

Figure 19. Relative differences of the new certified $n(^{234}U)/n(^{238}U)$ ratios for the IRMM-183-187 series from the previous certified ratios. The shown uncertainties include the uncertainties for both of them. The uncertainties are given with a coverage factor k=2.

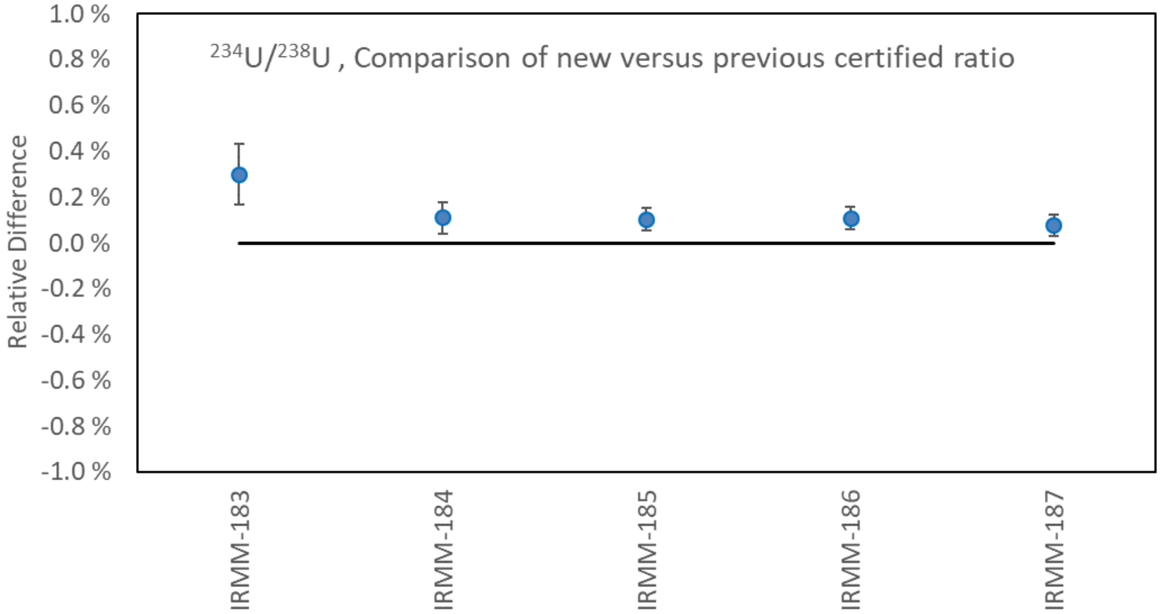
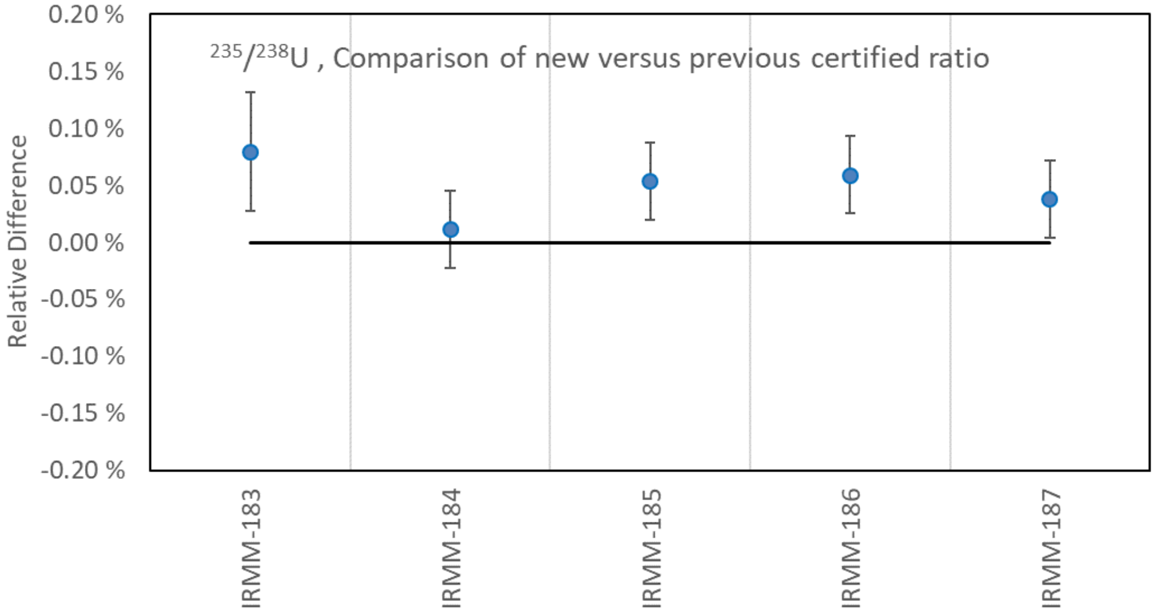


Figure 20. Relative differences of the new certified $n(^{235}U)/n(^{238}U)$ ratios for the IRMM-183-187 series from the previous certified ratios. The shown uncertainties include the uncertainties for both of them. The uncertainties are given with a coverage factor k=2.



For the $n(^{235}U)/n(^{238}U)$ ratio significant differences can be observed for the entire IRMM-183-187 series except for IRMM-184, as shown in Figure 20. The largest difference is observed for IRMM-183 (0.08 %), but also for IRMM-185, IRMM-186 and IRMM-187 the differences are at a remarkable level of about 0.04 % to 0.05 %. The reasons for these differences are not known, because not much detail is available about the previous certified $n(^{235}U)/n(^{238}U)$ ratios.

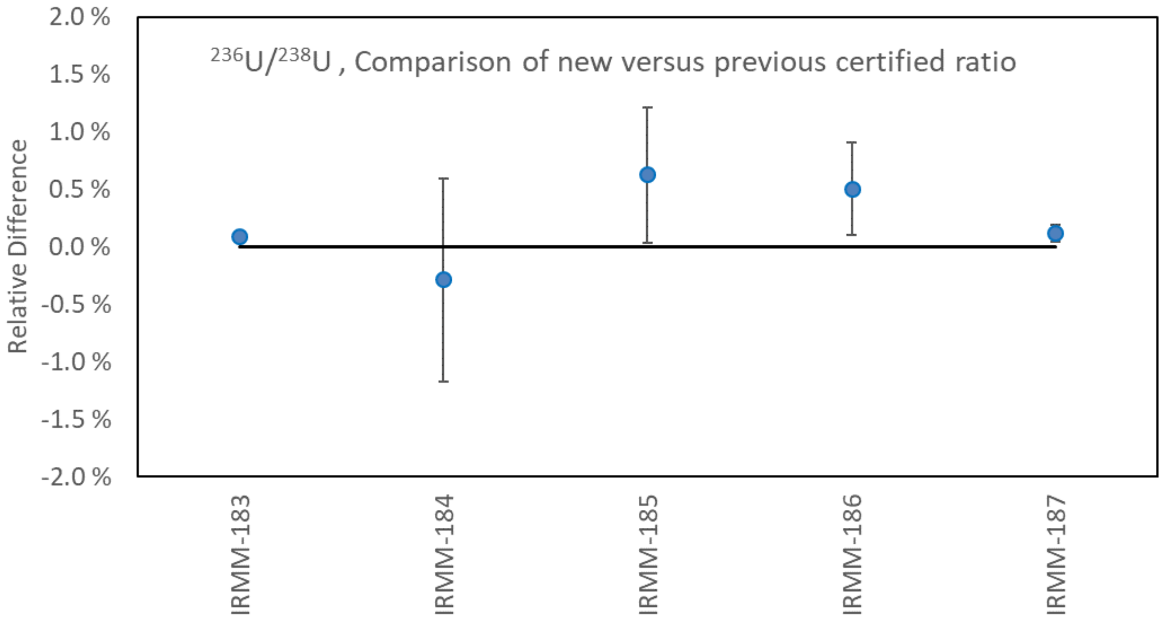
As shown in Figure 21, also for the $n(^{236}U)/n(^{238}U)$ ratio significant differences can be observed for the entire IRMM-183-187 series, except for the quite low ratio of 1.241×10^{-7} in case of IRMM-184.

The reason for the differences in the $n(^{236}U)/n(^{238}U)$ ratios in case of IRMM-183 and IRMM-187, measured by Faraday cups both in 2004 [4] and for the re-certification is the change in the peak tailing correction as explained above for the $n(^{234}U)/n(^{238}U)$ ratios.

The reason for the difference in the $n(^{236}U)/n(^{238}U)$ ratio in case of IRMM-185 measured using the SEM both in 2004 and for the re-certification might be difficulties with the SEM calibration, tailing and background corrections. The same might also apply to the $n(^{236}U)/n(^{238}U)$ ratio of IRMM-186, which was measured using the SEM in 2004 but using a Faraday cup for the re-certification.

However, for this project the validity of the SEM calibration was also verified by QC measurements of IRMM-187 with $n(^{236}U)/n(^{238}U)$ ratios of about 7×10^{-5} within the overlap region between SEM and Faraday cup detection. Furthermore, the new certified $n(^{236}U)/n(^{238}U)$ ratios for IRMM-185 and IRMM-186 have been verified successfully by external laboratories as shown in Figure 16 and Figure 17 and can therefore be relied on.

Figure 21. Relative differences of the new certified $n(^{236}U)/n(^{238}U)$ ratios for the IRMM-183-187 series from the previous certified ratios. The shown uncertainties include the uncertainties for both of them. The uncertainties are given with a coverage factor k=2.



7 Homogeneity Assessment

A key requirement for any reference material aliquoted into units is equivalence between those units. 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 was considered unnecessary due to the homogeneous nature of the isotopic composition of the uranium nitrate solutions. Evidence for this is presented in the context of the certification of the series IRMM-019-029 and IRMM-2019-2029, in which the uncertainty contribution from the homogeneity test was demonstrated at the level of the measurement uncertainty and the expected trends with the values of the isotope ratios [10, 11]. The analyses performed for the IRMM-183-187 series for this study, including the characterization measurements by JRC-G2 as well as the verification measurements by IAEA, ORNL and CEA, did not show any statistically significant difference between ampoules for the major or minor ratios.

8 Stability Assessment

8.1 Short-term stability study

The IRMM-183-187 series of reference material consists of uranium nitrate solution isotopic reference materials in HNO_3 (c = 1 mol / l) contained in flame sealed quartz ampoules from earlier (1990s). 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, respecting fissile and radioactive material transport regulations.

8.2 Long-term stability study

A long term stability assessment is considered to be unnecessary, due to the stable nature of the uranium isotope ratios in uranium nitrate solutions. This has been demonstrated in the course of the certification of the IRMM-019-029 and IRMM-2019-2029 series [10, 11], where no significant trends were observed for isotope ratios over a time period of 10 years. This was also shown in the QC chart for the major ratio $n(^{235}U)/n(^{238}U)$ of IRMM-184 in Figure 2. Therefore, the validity of the new certificates for the IRMM-183-187 series is given for 10 years.

9 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 3), potential within-unit and between-unit inhomogeneities, combined and expressed as u_{hom} (section 7), potential degradation during transport, u_{trn} (section 8.1), and potential degradation during storage, u_{lts} (section 8.2). 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 before in sections 7 and 8, no assessment of the homogeneity nor of the stability were deemed necessary for the highly enriched uranium nitrate solutions. Therefore the uncertainties u_{hom} , u_{trn} and u_{lts} are considered 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 of the IRMM-183-187 series.

9.1 Uranium isotope amount ratios, isotope amount fractions, isotope mass fractions and molar mass

The results of the characterization of the uranium isotope amount ratios for the IRMM-183-187 series of reference materials are presented within section 3 in Table 1. These results are assigned as the certified values. The uranium isotope amount ratios are given in mol / mol.

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:

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

Moreover, the molar mass of the uranium in the material 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) has been taken from the most recent atomic mass evaluation (AME 2020, [18]).

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 in the material:

$$\boldsymbol{m}({}^{x}\mathbf{U})/\boldsymbol{m}(\mathbf{U}) = \frac{\boldsymbol{n}\left({}^{x=(234:235:236:238)}\mathbf{U}\right)/\boldsymbol{n}(\mathbf{U}) \cdot \boldsymbol{M}({}^{x}\mathbf{U})}{\boldsymbol{M}(\mathbf{U})} \qquad \text{Equation 4}$$

The certified and calculated derived values related to the uranium isotopic composition for the IRMM-183-187 series are summarised in Table 4. The calculation of the certified isotope amount fractions, isotope mass fractions and the molar mass is shown in Annex 3 to Annex 7.

Table 4. Certified Values for IRMM-183-187 series

	$n(^{234}U)/n(^{238}U)$	$n(^{235}U)/n(^{238}U)$	$n(^{236}U)/n(^{238}U)$	
IRMM-183	0.000019814 (14)	0.00321826 (52)	0.000148492 (30)	
IRMM-184	0.000053196 (16)	0.0072631 (11)	0.00000012410 (96)	
IRMM-185	0.000179659 (39)	0.0200659 (32)	0.000002907 (12)	
IRMM-186	0.000293966 (64)	0.0307894 (48)	0.000033388 (34)	
IRMM-187	0.000387298 (91)	0.0473430 (75)	0.000072049 (38)	
	$n(^{234}U)/n(U)$	$n(^{235}U)/n(U)$	$n(^{236}U)/n(U)$	$n(^{238}U)/n(U)$
IRMM-183	0.000019747 (13)	0.00320739 (51)	0.000147991 (30)	0.99662487 (51)
IRMM-184	0.000052810 (15)	0.0072103 (11)	0.00000012320 (95)	0.9927368 (11)
IRMM-185	0.000176093 (38)	0.0196677 (30)	0.000002849 (12)	0.9801534 (30)
IRMM-186	0.000285094 (62)	0.0298602 (46)	0.000032380 (33)	0.9698223 (46)
IRMM-187	0.000369628 (87)	0.0451831 (68)	0.000068762 (36)	0.9543785 (68)
	$m(^{234}U)/m(U)$	$m(^{235}U)/m(U)$	$m(^{236}U)/m(U)$	$m(^{238}U)/m(U)$
IRMM-183	0.000019415 (13)	0.00316701 (51)	0.000146750 (30)	0.99666682 (51)
IRMM-184	0.000051925 (15)	0.0071199 (11)	0.00000012217 (95)	0.9928281 (11)
IRMM-185	0.000173171 (37)	0.0194242 (30)	0.000002826 (11)	0.9803998 (30)
IRMM-186	0.000280399 (61)	0.0294943 (45)	0.000032120 (33)	0.9701932 (45)
IRMM-187	0.000363612 (86)	0.0446382 (67)	0.000068222 (36)	0.9549300 (67)
	Molar Mass			
IRMM-183	238.0407668(35)			
IRMM-184	238.0288945(47)			
IRMM-185	237.9909371(97)			
IRMM-186	237.959793(14)			
IRMM-187	237.913308(21)			

10 Metrological traceability and commutability

10.1 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 the IRMM-183-187 series are traceable to the International SI via the certified $n(^{233}U)/n(^{236}U)$ isotope ratio of the gravimetrically prepared certified reference double spike material IRMM-3636a.

10.2 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 [19] 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 measurement community as calibrant, QC sample and reference material mainly for mass spectrometry analysis.

11 Instructions for use

11.1 Safety information

The IRMM-183-187 series reference material 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.

11.2 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.

11.3 Use of the certified values

The main purpose of these materials and their certified values is to calibrate and to assess method performance for mass spectrometry, i.e. for checking accuracy of analytical results and for calibration.

11.4 Use as a calibrant (*i.e.* reference material used for calibration purposes)

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

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 [20]:

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 %.

11.5 Use in quality control charts

The materials can also 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.

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List of abbreviations and definitions

ASTM	American Society for Testing and Materials
CRM	Certified reference material
DS	Double Spike
EC	European Commission
IAEA	International Atomic Energy Agency
JRC	Joint Research Centre
k	Coverage factor
M	Molar mass
MTE	Modified total evaporation
NBL	New Brunswick Laboratory Program Office
SEM	Secondary Electron Multiplier
SGAS	Safeguards Analytical Services of the IAEA
SI	International system of units
TIMS	Thermal ionisation mass spectrometry
U	Expanded 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)
UF_6	Uranium hexafluoride
VIM	International vocabulary on metrology

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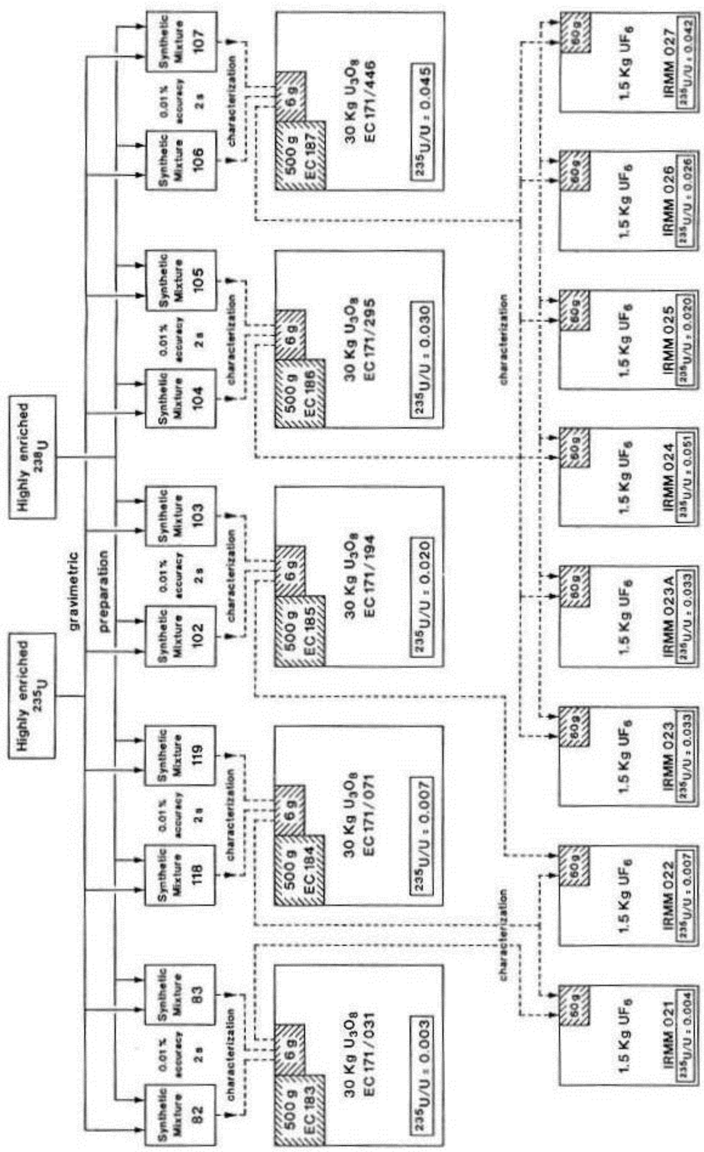
Annexes

Annex 1. Preparation of IRMM-183-187 Series

The following materials were used in the preparation of the IRMM-183-187 series:

1. Set of two "starting materials", highly enriched in ^{235}U and ^{238}U
2. Set of 10 gravimetrically prepared mixtures, subsequently fluorinated
3. IRMM-171 series (labelled EC171 here), 30 kg of oxides (U_3O_8), 6 g were fluorinated and characterised by GSMS using the set of fluorinated 10 mixtures
4. IRMM-183 – IRMM-187 series, uranium nitrate solutions (labelled EC183 – EC187 here), was prepared by dissolving 500g of IRMM-171 series
5. IRMM-021 – IRMM-027 (later extended towards IRMM-019 – IRMM-029), characterised by GSMS using fluorinated IRMM-171 series



Annex 2. Certificates of reference materials IRMM-3636a, IRMM-074 and 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 – 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

Signed: ______________ 1 8 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

1 8 SEP. 2019

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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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Directorate G – Nuclear Safety and Security
G.2 – Standards for Nuclear Safety, Security and Safeguards Unit
Retieseweg 111, B - 2440 Geel (Belgium)

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CERTIFIED REFERENCE MATERIAL
IRMM – 074

CERTIFICATE OF ANALYSIS

Uranium in nitric acid solution

Code number	Certified isotope amount ratios [1] [mol/mol]		
	$n(^{233}U)/n(^{235}U)$	$n(^{233}U)/n(^{238}U)$	$n(^{235}U)/n(^{238}U)$
	U = 0.025 % (relative) [2]	U = 0.025 % (relative) [2]	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
Directorate G – Nuclear Safety and Security
G.2 – Standards for Nuclear Safety, Security and Safeguards Unit
Retieseweg 111
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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Directorate G – Nuclear Safety and Security
G.2 – Standards for Nuclear Safety, Security and Safeguards Unit
Retieseweg 111, B - 2440 Geel (Belgium)

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CERTIFIED REFERENCE MATERIAL

IRMM-075

URANIUM NITRATE SOLUTION		
	Isotope amount ratio $n(^{236}U)/n(^{238}U)$	
	Certified value [1)] [mol/mol]	Uncertainty [2)] [mol/mol]
IRMM-075/1	1.04433×10^{-4}	3.7×10^{-8}
IRMM-075/2	1.14160×10^{-4}	4.0×10^{-9}
IRMM-075/3	1.04093×10^{-6}	3.6×10^{-10}
IRMM-075/4	1.13742×10^{-7}	4.0×10^{-11}
IRMM-075/5	1.06519×10^{-8}	7.5×10^{-12}
IRMM-075/6	1.0885×10^{-9}	6.3×10^{-12}

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

Due to the homogenous nature of the material, there is no minimum sample intake to be considered.

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

This certificate is a revision of the revised certificate issued in 2019, which is based on the original certificate of 2007.

Geel,

Signed: 3-11-2021

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

DESCRIPTION OF THE MATERIAL

IRMM-075 is an uranyl nitrate solution isotope reference material 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 nitric acid solution in a sealed quartz ampoule. The concentration of the nitric acid is 1 mol/L.

ANALYTICAL METHODS USED FOR CHARACTERIZATION

The certified values were established by gravimetric mixing of highly enriched ^{236}U and ^{238}U starting materials and verified by thermal ionisation mass spectrometry (TIMS).

SAFETY INFORMATION

The IRMM-075 contains radioactive material. The ampoule 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 reference material is intended for the verification and correction of non-linearities of the entire mass spectrometer measurement system.

STORAGE

The ampoule should be stored at room temperature.

However, the European Commission cannot be held responsible for changes that happen during storage of the material at the customer's premises, especially for opened samples.

LEGAL NOTICE

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(b) assume any liability with respect to, or for damages resulting from, the use of any information, material, apparatus, method or process disclosed in this document save for loss or damage arising solely and directly from the negligence of Joint Research Centre of the European Commission.

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 the Joint Research Centre on request.

European Commission – Joint Research Centre
Directorate G – Nuclear Safety and Security
Retieseweg 111, B - 2440 Geel (Belgium)

Telephone: +32-(0)14-571-855
Email: JRC-GEE-RM-NUCLEAR-SALES@ec.europa.eu

Annex 3. Calculation of isotope abundance fractions, isotope mass fractions and molar mass for IRMM-183.

IRMM-183

Measurements performed using TIMS/MTE and TIMS/DS, according to Method Working Instruction: IMS-JRC.G-C1.1-WIN-0059 v3.0

All results of mass spectrometric measurements for this request are stored in "U:\Nuclear Safeguards\Nuclear RE$_{\text{CERTIFICATION\textbackslash Measurement}}$ Results\T210704 MTE NORM DS - IRMM-183 - Turret-8 - all combined - Rev 01.xls"

So this is from the XLS file ! The original XLSM file had to be stored as XLS for GUMWB to be able to use the data directly.

Model Equation:

{-- final ratios of sample--}

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

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	g/mol	molar mass of U
f_{233U}	mol/mol	isotope amount fraction of ^{233}U in U
f_{234U}	mol/mol	isotope amount fraction of ^{234}U in U
f_{235U}	mol/mol	isotope amount fraction of ^{235}U in U
f_{236U}	mol/mol	isotope amount fraction of ^{236}U in U
f_{238U}	mol/mol	isotope amount fraction of ^{238}U in U
w_{233U}	mol/mol	isotope mass fraction of ^{233}U in U
w_{234U}	mol/mol	isotope mass fraction of ^{234}U in U
w_{235U}	mol/mol	isotope mass fraction of ^{235}U in U
w_{236U}	mol/mol	isotope mass fraction of ^{236}U in U

Quantity	Unit	Definition
w_{238U}	mol/mol	isotope mass fraction of ^{238}U in U
M_{233U}	g/mol	atomic mass for ^{233}U
M_{234U}	g/mol	atomic mass for ^{234}U
M_{235U}	g/mol	atomic mass for ^{235}U
M_{236U}	g/mol	atomic mass for ^{236}U
M_{238U}	g/mol	atomic mass for ^{238}U
ΣR_U	mol/mol	sum of isotope ratios for U

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

$R_{234/238U}$:
Import from Excel
Filename: T210704 MTE NORM DS - IRMM-183 - Turret-8 - all combined - Rev 01.xls
Worksheet: Summary-calc
Value Cell: H176 = $19.81377 \cdot 10^{-6}$ mol/mol
Standarduncertainty Cell: H177 = $6.77 \cdot 10^{-9}$ mol/mol
Degrees of Freedom Cell: E51 = 10

$R_{235/238U}$:
Import from Excel
Filename: T210704 MTE NORM DS - IRMM-183 - Turret-8 - all combined - Rev 01.xls
Worksheet: Summary-calc
Value Cell: G176 = $3.218256 \cdot 10^{-3}$ mol/mol
Standarduncertainty Cell: G177 = $259 \cdot 10^{-9}$ mol/mol
Degrees of Freedom Cell: E51 = 10

$R_{236/238U}$:
Import from Excel
Filename: T210704 MTE NORM DS - IRMM-183 - Turret-8 - all combined - Rev 01.xlsm
Worksheet: Summary-calc
Value Cell: I176 = $148.4920 \cdot 10^{-6}$ mol/mol
Standarduncertainty Cell: I177 = $15.0 \cdot 10^{-9}$ mol/mol
Degrees of Freedom Cell: E51 = 10

M_{233U}:
Type B normal distribution
Value: 233.039627 g/mol
Expanded Uncertainty: 0.000003 g/mol
Coverage Factor: 1.0

Meng Wang et al (2021), The AME 2020 atomic mass evaluation, Chinese Phys. C 45 030003

M_{234U}:
Type B normal distribution
Value: 234.0409503 g/mol
Expanded Uncertainty: 0.0000024 g/mol
Coverage Factor: 2

Meng Wang et al (2021), The AME 2020 atomic mass evaluation, Chinese Phys. C 45 030003

M_{235U}: Type B normal distribution
Value: 235.0439281 g/mol
Expanded Uncertainty: 0.0000024 g/mol
Coverage Factor: 2.0

Meng Wang et al (2021), The AME 2020 atomic mass evaluation, Chinese Phys. C 45 030003

M_{236U}: Type B normal distribution
Value: 236.0455661 g/mol
Expanded Uncertainty: 0.0000024 g/mol
Coverage Factor: 2.0

Meng Wang et al (2021), The AME 2020 atomic mass evaluation, Chinese Phys. C 45 030003

M_{238U}: Type B normal distribution
Value: 238.0507869 g/mol
Expanded Uncertainty: 0.0000032 g/mol
Coverage Factor: 2.0

Meng Wang et al (2021), The AME 2020 atomic mass evaluation, Chinese Phys. C 45 030003

Interim Results:

Quantity	Value	Standard Uncertainty
ΣR_U	1.003386561 mol/mol	$259 \cdot 10^{-9}$ mol/mol

Results:

Quantity	Value	Expanded Uncertainty	Coverage factor	Coverage
M_U	238.0407668 g/mol	$3.5 \cdot 10^{-6}$ g/mol	2.00	manual
f_{234U}	$19.747 \cdot 10^{-6}$ mol/mol	$13 \cdot 10^{-9}$ mol/mol	2.00	manual
f_{235U}	$3.20739 \cdot 10^{-3}$ mol/mol	$510 \cdot 10^{-9}$ mol/mol	2.00	manual
f_{236U}	$147.991 \cdot 10^{-6}$ mol/mol	$30 \cdot 10^{-9}$ mol/mol	2.00	manual
f_{238U}	0.99662487 mol/mol	$510 \cdot 10^{-9}$ mol/mol	2.00	manual
w_{234U}	$19.415 \cdot 10^{-6}$ mol/mol	$13 \cdot 10^{-9}$ mol/mol	2.00	manual
w_{235U}	$3.16701 \cdot 10^{-3}$ mol/mol	$510 \cdot 10^{-9}$ mol/mol	2.00	manual
w_{236U}	$146.750 \cdot 10^{-6}$ mol/mol	$30 \cdot 10^{-9}$ mol/mol	2.00	manual
w_{238U}	0.99666682 mol/mol	$510 \cdot 10^{-9}$ mol/mol	2.00	manual

Annex 4. Calculation of isotope abundance fractions, isotope mass fractions and molar mass for IRMM-184

IRMM-184

Measurements performed using TIMS/MTE and TIMS/DS, according to Method Working Instruction: IMS-JRC.G-C1.1-WIN-0059 v3.0

All results of mass spectrometric measurements for this request are stored in "U:\Nuclear Safeguards\Nuclear RE$_{\text{CERTIFICATION\textbackslash Measurement}}$ Results\T210715 MTE NORM DS - IRMM-184 - SEM - Turret-11 - Rev 01.xls"

So this is from the XLS file ! The original XLSM file had to be stored as XLS for GUMWB to be able to use the data directly.

Model Equation:

{-- final ratios of sample---}

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

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

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

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

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

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

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

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

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

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

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

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

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	g/mol	molar mass of U
f_{233U}	mol/mol	isotope amount fraction of ^{233}U in U
f_{234U}	mol/mol	isotope amount fraction of ^{234}U in U
f_{235U}	mol/mol	isotope amount fraction of ^{235}U in U
f_{236U}	mol/mol	isotope amount fraction of ^{236}U in U
f_{238U}	mol/mol	isotope amount fraction of ^{238}U in U
w_{233U}	mol/mol	isotope mass fraction of ^{233}U in U
w_{234U}	mol/mol	isotope mass fraction of ^{234}U in U
w_{235U}	mol/mol	isotope mass fraction of ^{235}U in U
w_{236U}	mol/mol	isotope mass fraction of ^{236}U in U

Quantity	Unit	Definition
w_{238U}	mol/mol	isotope mass fraction of ^{238}U in U
M_{233U}	g/mol	atomic mass for ^{233}U
M_{234U}	g/mol	atomic mass for ^{234}U
M_{235U}	g/mol	atomic mass for ^{235}U
M_{236U}	g/mol	atomic mass for ^{236}U
M_{238U}	g/mol	atomic mass for ^{238}U
ΣR_U	mol/mol	sum of isotope ratios for U

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

$R_{234/238U}$:
Import from Excel
Filename: T210715 MTE NORM DS - IRMM-184 - SEM - Turret-11 - Rev 01.xls
Worksheet: Summary-calc
Value Cell: H176 = $53.19598 \cdot 10^{-6}$ mol/mol
Standarduncertainty Cell: H177 = $7.80 \cdot 10^{-9}$ mol/mol
Degrees of Freedom Cell: E51 = 7

$R_{235/238U}$:
Import from Excel
Filename: T210715 MTE NORM DS - IRMM-184 - SEM - Turret-11 - Rev 01.xls
Worksheet: Summary-calc
Value Cell: G176 = $7.263063 \cdot 10^{-3}$ mol/mol
Standarduncertainty Cell: G177 = $574 \cdot 10^{-9}$ mol/mol
Degrees of Freedom Cell: E51 = 7

$R_{236/238U}$:
Import from Excel
Filename: T210715 MTE NORM DS - IRMM-184 - SEM - Turret-11 - Rev 01.xls
Worksheet: Summary-calc
Value Cell: I176 = $124.102 \cdot 10^{-9}$ mol/mol
Standarduncertainty Cell: I177 = $480 \cdot 10^{-12}$ mol/mol
Degrees of Freedom Cell: E51 = 7

M_{233U}:
Type B normal distribution
Value: 233.039627 g/mol
Expanded Uncertainty: 0.000003 g/mol
Coverage Factor: 1.0

Meng Wang et al (2021), The AME 2020 atomic mass evaluation, Chinese Phys. C 45 030003

M_{234U}:
Type B normal distribution
Value: 234.0409503 g/mol
Expanded Uncertainty: 0.0000024 g/mol
Coverage Factor: 2

Meng Wang et al (2021), The AME 2020 atomic mass evaluation, Chinese Phys. C 45 030003

M_{235U}:
Type B normal distribution
Value: 235.0439281 g/mol
Expanded Uncertainty: 0.0000024 g/mol
Coverage Factor: 2.0

Meng Wang et al (2021), The AME 2020 atomic mass evaluation, Chinese Phys. C 45 030003

M_{236U}: Type B normal distribution
Value: 236.0455661 g/mol
Expanded Uncertainty: 0.0000024 g/mol
Coverage Factor: 2.0

Meng Wang et al (2021), The AME 2020 atomic mass evaluation, Chinese Phys. C 45 030003

M_{238U}: Type B normal distribution
Value: 238.0507869 g/mol
Expanded Uncertainty: 0.0000032 g/mol
Coverage Factor: 2.0

Meng Wang et al (2021), The AME 2020 atomic mass evaluation, Chinese Phys. C 45 030003

Interim Results:

Quantity	Value	Standard Uncertainty
ΣR_U	1.007316383 mol/mol	$574 \cdot 10^{-9}$ mol/mol

Results:

Quantity	Value	Expanded Uncertainty	Coverage factor	Coverage
M_U	238.0288945 g/mol	$4.7 \cdot 10^{-6}$ g/mol	2.00	manual
f_{234U}	$52.810 \cdot 10^{-6}$ mol/mol	$15 \cdot 10^{-9}$ mol/mol	2.00	manual
f_{235U}	$7.2103 \cdot 10^{-3}$ mol/mol	$1.1 \cdot 10^{-6}$ mol/mol	2.00	manual
f_{236U}	$123.20 \cdot 10^{-9}$ mol/mol	$950 \cdot 10^{-12}$ mol/mol	2.00	manual
f_{238U}	0.9927368 mol/mol	$1.1 \cdot 10^{-6}$ mol/mol	2.00	manual
w_{234U}	$51.925 \cdot 10^{-6}$ mol/mol	$15 \cdot 10^{-9}$ mol/mol	2.00	manual
w_{235U}	$7.1199 \cdot 10^{-3}$ mol/mol	$1.1 \cdot 10^{-6}$ mol/mol	2.00	manual
w_{236U}	$122.17 \cdot 10^{-9}$ mol/mol	$950 \cdot 10^{-12}$ mol/mol	2.00	manual
w_{238U}	0.9928281 mol/mol	$1.1 \cdot 10^{-6}$ mol/mol	2.00	manual

Annex 5. Calculation of isotope abundance fractions, isotope mass fractions and molar mass for IRMM-185

IRMM-185

Measurements performed using TIMS/MTE and TIMS/DS, according to Method Working Instruction: IMS-JRC.G-C1.1-WIN-0059 v3.0

All results of mass spectrometric measurements for this request are stored in "T210920 MTE-NORM-DS - IRMM- 185 - SEM - Turret-14a - all combined - Rev 01.xls"

So this is from the XLS file ! The original XLSM file had to be stored as XLS for GUMWB to be able to use the data directly.

Model Equation:

{-- final ratios of sample--}

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

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

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

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

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

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

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

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

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

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

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

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

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	g/mol	molar mass of U
f_{233U}	mol/mol	isotope amount fraction of ^{233}U in U
f_{234U}	mol/mol	isotope amount fraction of ^{234}U in U
f_{235U}	mol/mol	isotope amount fraction of ^{235}U in U
f_{236U}	mol/mol	isotope amount fraction of ^{236}U in U
f_{238U}	mol/mol	isotope amount fraction of ^{238}U in U
w_{233U}	mol/mol	isotope mass fraction of ^{233}U in U
w_{234U}	mol/mol	isotope mass fraction of ^{234}U in U
w_{235U}	mol/mol	isotope mass fraction of ^{235}U in U
w_{236U}	mol/mol	isotope mass fraction of ^{236}U in U

Quantity	Unit	Definition
w_{238U}	mol/mol	isotope mass fraction of ^{238}U in U
M_{233U}	g/mol	atomic mass for ^{233}U
M_{234U}	g/mol	atomic mass for ^{234}U
M_{235U}	g/mol	atomic mass for ^{235}U
M_{236U}	g/mol	atomic mass for ^{236}U
M_{238U}	g/mol	atomic mass for ^{238}U
ΣR_U	mol/mol	sum of isotope ratios for U

$R_{233/238U}$:

Type B normal distribution
Value: 0 mol/mol
Expanded Uncertainty: 0 mol/mol
Coverage Factor: 2

$R_{234/238U}$:

Import from Excel
Filename: T210920 MTE-NORM-DS - IRMM-185 - SEM - Turret-14a - all combined - Rev 01.xls
Worksheet: Summary-calc
Value Cell: H176 = $179.6589 \cdot 10^{-6}$ mol/mol
Standarduncertainty Cell: H177 = $19.4 \cdot 10^{-9}$ mol/mol
Degrees of Freedom Cell: E51 = 8

$R_{235/238U}$:

Import from Excel
Filename: T210920 MTE-NORM-DS - IRMM-185 - SEM - Turret-14a - all combined - Rev 01.xls
Worksheet: Summary-calc
Value Cell: G176 = 0.02006594 mol/mol
Standarduncertainty Cell: G177 = $1.58 \cdot 10^{-6}$ mol/mol
Degrees of Freedom Cell: E51 = 8

$R_{236/238U}$:

Import from Excel
Filename: T210920 MTE-NORM-DS - IRMM-185 - SEM - Turret-14a - all combined - Rev 01.xls
Worksheet: Summary-calc
Value Cell: I176 = $2.90709 \cdot 10^{-6}$ mol/mol
Standarduncertainty Cell: I177 = $5.88 \cdot 10^{-9}$ mol/mol
Degrees of Freedom Cell: E51 = 8

M_{233U}:

Type B normal distribution
Value: 233.039627 g/mol
Expanded Uncertainty: 0.000003 g/mol
Coverage Factor: 1.0

Meng Wang et al (2021), The AME 2020 atomic mass evaluation, Chinese Phys. C 45 030003

M_{234U}:

Type B normal distribution
Value: 234.0409503 g/mol
Expanded Uncertainty: 0.0000024 g/mol
Coverage Factor: 2

Meng Wang et al (2021), The AME 2020 atomic mass evaluation, Chinese Phys. C 45 030003

M_{235U}: Type B normal distribution
Value: 235.0439281 g/mol
Expanded Uncertainty: 0.0000024 g/mol
Coverage Factor: 2.0

Meng Wang et al (2021), The AME 2020 atomic mass evaluation, Chinese Phys. C 45 030003

M_{236U}: Type B normal distribution
Value: 236.0455661 g/mol
Expanded Uncertainty: 0.0000024 g/mol
Coverage Factor: 2.0

Meng Wang et al (2021), The AME 2020 atomic mass evaluation, Chinese Phys. C 45 030003

M_{238U}: Type B normal distribution
Value: 238.0507869 g/mol
Expanded Uncertainty: 0.0000032 g/mol
Coverage Factor: 2.0

Meng Wang et al (2021), The AME 2020 atomic mass evaluation, Chinese Phys. C 45 030003

Interim Results:

Quantity	Value	Standard Uncertainty
ΣR_U	1.02024851 mol/mol	$1.58 \cdot 10^{-6}$ mol/mol

Results:

Quantity	Value	Expanded Uncertainty	Coverage factor	Coverage
M_U	237.9909371 g/mol	$9.7 \cdot 10^{-6}$ g/mol	2.00	manual
f_{234U}	$176.093 \cdot 10^{-6}$ mol/mol	$38 \cdot 10^{-9}$ mol/mol	2.00	manual
f_{235U}	0.0196677 mol/mol	$3.0 \cdot 10^{-6}$ mol/mol	2.00	manual
f_{236U}	$2.849 \cdot 10^{-6}$ mol/mol	$12 \cdot 10^{-9}$ mol/mol	2.00	manual
f_{238U}	0.9801534 mol/mol	$3.0 \cdot 10^{-6}$ mol/mol	2.00	manual
w_{234U}	$173.171 \cdot 10^{-6}$ mol/mol	$37 \cdot 10^{-9}$ mol/mol	2.00	manual
w_{235U}	0.0194242 mol/mol	$3.0 \cdot 10^{-6}$ mol/mol	2.00	manual
w_{236U}	$2.826 \cdot 10^{-6}$ mol/mol	$11 \cdot 10^{-9}$ mol/mol	2.00	manual
w_{238U}	0.9803998 mol/mol	$3.0 \cdot 10^{-6}$ mol/mol	2.00	manual

Annex 6. Calculation of isotope abundance fractions, isotope mass fractions and molar mass for IRMM-186

	IRMM-186	

IRMM-186

Measurements performed using TIMS/MTE and TIMS/DS, according to Method Working Instruction: IMS-JRC.G-C1.1-WIN-0059 v3.0

All results of mass spectrometric measurements for this request are stored in "T210824 MTE-NORM-DS - IRMM- 186 - 1E13 Ohm - Turret-12 - all combined - Rev 01.xls"

So this is from the XLS file ! The original XLSM file had to be stored as XLS for GUMWB to be able to use the data directly.

Model Equation:

{--- final ratios of sample---}

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

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

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

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

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

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

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

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

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

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

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

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

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	g/mol	molar mass of U
f_{233U}	mol/mol	isotope amount fraction of ^{233}U in U
f_{234U}	mol/mol	isotope amount fraction of ^{234}U in U
f_{235U}	mol/mol	isotope amount fraction of ^{235}U in U
f_{236U}	mol/mol	isotope amount fraction of ^{236}U in U
f_{238U}	mol/mol	isotope amount fraction of ^{238}U in U
w_{233U}	mol/mol	isotope mass fraction of ^{233}U in U
w_{234U}	mol/mol	isotope mass fraction of ^{234}U in U
w_{235U}	mol/mol	isotope mass fraction of ^{235}U in U
w_{236U}	mol/mol	isotope mass fraction of ^{236}U in U

Date: 03/30/2022	File: IRMM-186.smu	Page 1 of 3

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Quantity	Unit	Definition
w_{238U}	mol/mol	isotope mass fraction of ^{238}U in U
M_{233U}	g/mol	atomic mass for ^{233}U
M_{234U}	g/mol	atomic mass for ^{234}U
M_{235U}	g/mol	atomic mass for ^{235}U
M_{236U}	g/mol	atomic mass for ^{236}U
M_{238U}	g/mol	atomic mass for ^{238}U
ΣR_U	mol/mol	sum of isotope ratios for U

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

$R_{234/238U}$:
Import from Excel
Filename: T210824 MTE-NORM-DS - IRMM-186 - 1E13 Ohm - Turret-12 - all combined - Rev 01.xls
Worksheet: Summary-calc
Value Cell: H176 = $293.9655 \cdot 10^{-6}$ mol/mol
Standarduncertainty Cell: H177 = $31.8 \cdot 10^{-9}$ mol/mol
Degrees of Freedom Cell: E51 = 10

$R_{235/238U}$:
Import from Excel
Filename: T210824 MTE-NORM-DS - IRMM-186 - 1E13 Ohm - Turret-12 - all combined - Rev 01.xls
Worksheet: Summary-calc
Value Cell: G176 = 0.03078936 mol/mol
Standarduncertainty Cell: G177 = $2.42 \cdot 10^{-6}$ mol/mol
Degrees of Freedom Cell: E51 = 10

$R_{236/238U}$:
Import from Excel
Filename: T210824 MTE-NORM-DS - IRMM-186 - 1E13 Ohm - Turret-12 - all combined - Rev 01.xls
Worksheet: Summary-calc
Value Cell: I176 = $33.3876 \cdot 10^{-6}$ mol/mol
Standarduncertainty Cell: I177 = $17.1 \cdot 10^{-9}$ mol/mol
Degrees of Freedom Cell: E51 = 10

M_{233U}:
Type B normal distribution
Value: 233.039627 g/mol
Expanded Uncertainty: 0.000003 g/mol
Coverage Factor: 1.0

Meng Wang et al (2021), The AME 2020 atomic mass evaluation, Chinese Phys. C 45 030003

M_{234U}:
Type B normal distribution
Value: 234.0409503 g/mol
Expanded Uncertainty: 0.0000024 g/mol
Coverage Factor: 2

Meng Wang et al (2021), The AME 2020 atomic mass evaluation, Chinese Phys. C 45 030003

M_{235U}: Type B normal distribution
Value: 235.0439281 g/mol
Expanded Uncertainty: 0.0000024 g/mol
Coverage Factor: 2.0

Meng Wang et al (2021), The AME 2020 atomic mass evaluation, Chinese Phys. C 45 030003

M_{236U}: Type B normal distribution
Value: 236.0455661 g/mol
Expanded Uncertainty: 0.0000024 g/mol
Coverage Factor: 2.0

Meng Wang et al (2021), The AME 2020 atomic mass evaluation, Chinese Phys. C 45 030003

M_{238U}: Type B normal distribution
Value: 238.0507869 g/mol
Expanded Uncertainty: 0.0000032 g/mol
Coverage Factor: 2.0

Meng Wang et al (2021), The AME 2020 atomic mass evaluation, Chinese Phys. C 45 030003

Interim Results:

Quantity	Value	Standard Uncertainty
ΣR_U	1.03111672 mol/mol	$2.42\cdot10^{-6}$ mol/mol

Results:

Quantity	Value	Expanded Uncertainty	Coverage factor	Coverage
M_U	237.959793 g/mol	$14\cdot10^{-6}$ g/mol	2.00	manual
f_{234U}	$285.094\cdot10^{-6}$ mol/mol	$62\cdot10^{-9}$ mol/mol	2.00	manual
f_{235U}	0.0298602 mol/mol	$4.6\cdot10^{-6}$ mol/mol	2.00	manual
f_{236U}	$32.380\cdot10^{-6}$ mol/mol	$33\cdot10^{-9}$ mol/mol	2.00	manual
f_{238U}	0.9698223 mol/mol	$4.6\cdot10^{-6}$ mol/mol	2.00	manual
w_{234U}	$280.399\cdot10^{-6}$ mol/mol	$61\cdot10^{-9}$ mol/mol	2.00	manual
w_{235U}	0.0294943 mol/mol	$4.5\cdot10^{-6}$ mol/mol	2.00	manual
w_{236U}	$32.120\cdot10^{-6}$ mol/mol	$33\cdot10^{-9}$ mol/mol	2.00	manual
w_{238U}	0.9701932 mol/mol	$4.5\cdot10^{-6}$ mol/mol	2.00	manual

Annex 7. Calculation of isotope abundance fractions, isotope mass fractions and molar mass for IRMM-187

IRMM-187

Measurements performed using TIMS/MTE and TIMS/DS, according to Method Working Instruction: IMS-JRC.G-C1.1-WIN-0059 v3.0

All results of mass spectrometric measurements for this request are stored in "T210906 MTE-NORM-DS - IRMM- 187 - 1E13 Ohm - Turret-13a - all combined - Rev 01.xls"

So this is from the XLS file ! The original XLSM file had to be stored as XLS for GUMWB to be able to use the data directly.

Model Equation:

{-- final ratios of sample---}

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

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

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

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

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

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

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

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

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

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

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

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

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	g/mol	molar mass of U
f_{233U}	mol/mol	isotope amount fraction of ^{233}U in U
f_{234U}	mol/mol	isotope amount fraction of ^{234}U in U
f_{235U}	mol/mol	isotope amount fraction of ^{235}U in U
f_{236U}	mol/mol	isotope amount fraction of ^{236}U in U
f_{238U}	mol/mol	isotope amount fraction of ^{238}U in U
w_{233U}	mol/mol	isotope mass fraction of ^{233}U in U
w_{234U}	mol/mol	isotope mass fraction of ^{234}U in U
w_{235U}	mol/mol	isotope mass fraction of ^{235}U in U
w_{236U}	mol/mol	isotope mass fraction of ^{236}U in U

Quantity	Unit	Definition
w_{238U}	mol/mol	isotope mass fraction of ^{238}U in U
M_{233U}	g/mol	atomic mass for ^{233}U
M_{234U}	g/mol	atomic mass for ^{234}U
M_{235U}	g/mol	atomic mass for ^{235}U
M_{236U}	g/mol	atomic mass for ^{236}U
M_{238U}	g/mol	atomic mass for ^{238}U
ΣR_U	mol/mol	sum of isotope ratios for U

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

$R_{234/238U}$:
Import from Excel
Filename: T210906 MTE-NORM-DS - IRMM-187 - 1E13 Ohm - Turret-13a - all combined - Rev 01.xls
Worksheet: Summary-calc
Value Cell: H176 = $387.2976 \cdot 10^{-6}$ mol/mol
Standarduncertainty Cell: H177 = $45.6 \cdot 10^{-9}$ mol/mol
Degrees of Freedom Cell: E51 = 11

$R_{235/238U}$:
Import from Excel
Filename: T210906 MTE-NORM-DS - IRMM-187 - 1E13 Ohm - Turret-13a - all combined - Rev 01.xls
Worksheet: Summary-calc
Value Cell: G176 = 0.04734297 mol/mol
Standarduncertainty Cell: G177 = $3.73 \cdot 10^{-6}$ mol/mol
Degrees of Freedom Cell: E51 = 11

$R_{236/238U}$:
Import from Excel
Filename: T210906 MTE-NORM-DS - IRMM-187 - 1E13 Ohm - Turret-13a - all combined - Rev 01.xls
Worksheet: Summary-calc
Value Cell: I176 = $72.0492 \cdot 10^{-6}$ mol/mol
Standarduncertainty Cell: I177 = $18.9 \cdot 10^{-9}$ mol/mol
Degrees of Freedom Cell: E51 = 11

M_{233U}:
Type B normal distribution
Value: 233.039627 g/mol
Expanded Uncertainty: 0.000003 g/mol
Coverage Factor: 1.0

Meng Wang et al (2021), The AME 2020 atomic mass evaluation, Chinese Phys. C 45 030003

M_{234U}:
Type B normal distribution
Value: 234.0409503 g/mol
Expanded Uncertainty: 0.0000024 g/mol
Coverage Factor: 2

Meng Wang et al (2021), The AME 2020 atomic mass evaluation, Chinese Phys. C 45 030003

M_{235U}: Type B normal distribution
Value: 235.0439281 g/mol
Expanded Uncertainty: 0.0000024 g/mol
Coverage Factor: 2.0

Meng Wang et al (2021), The AME 2020 atomic mass evaluation, Chinese Phys. C 45 030003

M_{236U}: Type B normal distribution
Value: 236.0455661 g/mol
Expanded Uncertainty: 0.0000024 g/mol
Coverage Factor: 2.0

Meng Wang et al (2021), The AME 2020 atomic mass evaluation, Chinese Phys. C 45 030003

M_{238U}: Type B normal distribution
Value: 238.0507869 g/mol
Expanded Uncertainty: 0.0000032 g/mol
Coverage Factor: 2.0

Meng Wang et al (2021), The AME 2020 atomic mass evaluation, Chinese Phys. C 45 030003

Interim Results:

Quantity	Value	Standard Uncertainty
ΣR_U	1.04780232 mol/mol	$3.74 \cdot 10^{-6}$ mol/mol

Results:

Quantity	Value	Expanded Uncertainty	Coverage factor	Coverage
M_U	237.913308 g/mol	$21 \cdot 10^{-6}$ g/mol	2.00	manual
f_{234U}	$369.628 \cdot 10^{-6}$ mol/mol	$87 \cdot 10^{-9}$ mol/mol	2.00	manual
f_{235U}	0.0451831 mol/mol	$6.8 \cdot 10^{-6}$ mol/mol	2.00	manual
f_{236U}	$68.762 \cdot 10^{-6}$ mol/mol	$36 \cdot 10^{-9}$ mol/mol	2.00	manual
f_{238U}	0.9543785 mol/mol	$6.8 \cdot 10^{-6}$ mol/mol	2.00	manual
w_{234U}	$363.612 \cdot 10^{-6}$ mol/mol	$86 \cdot 10^{-9}$ mol/mol	2.00	manual
w_{235U}	0.0446382 mol/mol	$6.7 \cdot 10^{-6}$ mol/mol	2.00	manual
w_{236U}	$68.222 \cdot 10^{-6}$ mol/mol	$36 \cdot 10^{-9}$ mol/mol	2.00	manual
w_{238U}	0.9549300 mol/mol	$6.7 \cdot 10^{-6}$ mol/mol	2.00	manual

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