European Commission

JRC REFERENCE MATERIALS REPORT

CERTIFICATION REPORT

Preparation and certification of a new batch of ^{242}Pu spike: IRMM-049e

Certified reference material for the ^{242}Pu amount content and Pu isotope amount ratios

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Abstract

This report describes the preparation and certification of the ^{242}Pu spike reference material IRMM-049e, applied for determination of plutonium content in nuclear materials by isotope dilution mass spectrometry (IDMS) in nuclear safeguards, nuclear security and in the industry. As the stock of IRMM-049d is close to exhaustion, it was decided to produce about 90 ampoules of a new spike certified reference material (CRM) IRMM-049e and, therefore to maintain the provision of the IRMM-049 series of spike CRMs.

IRMM-049e is the first of the IRMM-049 series produced in compliance with ISO 17034:2016.

The amount content of ^{242}Pu in IRMM-049e was characterised on 10 randomly stratified selected units by Isotope Dilution – Thermal Ionisation Mass Spectrometry (ID-TIMS) using the IRMM-1027o CRM as a spike. The plutonium isotope amount ratios were measured using the same 10 selected units by TIMS. The material was finally certified for the amount content of ^{242}Pu and the total Pu, the mass fractions of ^{242}Pu and total Pu, the Pu isotope amount ratios as well as the plutonium isotope abundances as amount and mass fractions, and for the molar mass of Pu in IRMM-049e.

The certified values were confirmed by ID-TIMS using the IRMM-086 (^{239}Pu spike) and verified against the reference value of the external certified test sample Pu EQRAIN-13 provided by CEA/CETAMA.

These verification studies were carried out in the frame of the on-going inter-calibration campaign using state-of-the art measurement procedures linking together different JRC plutonium spike reference materials and also external reference materials to underpin the confidence in the use of JRC isotopic plutonium reference materials for safeguards verification.

Between unit-homogeneity was quantified in accordance with ISO Guide 35:2006. No stability study was performed for this reference material since the general behaviour of this material is well known from past experience. However, a post-certification stability monitoring will be done every two years to control its stability after issuance of the certificate.

The uncertainties of the certified values were estimated in compliance with the Guide to the Expression of Uncertainty in Measurement (GUM) and include uncertainties related to characterisation and possible inhomogeneity.

The main purpose of this material is for use as a spike isotopic reference material for quantification of plutonium in an unknown nuclear sample. IRMM-049e is supplied in a screw-cap glass ampoule containing 10 mL nitric acid solution (c = 5 mol/L) with a certified plutonium (total Pu) mass fraction of (91.52 ± 0.12) µg/g and a certified ^{242}Pu amount content of (0.35828 ± 0.00045) µmol/g.

Because the IRMM-049e material is a true solution and as such, can be regarded as completely homogeneous, there is no minimum sample intake to be taken into account for the analysis.

The following values were assigned to IRMM-049e:

PLUTONIUM IN NITRIC ACID SOLUTION		
	Amount content	
	Certified value [1)] [μmol/g]	Uncertainty [2)] [μmol/g]
^{242}Pu	0.35828	0.00045
Pu	0.37826	0.00047
	Isotope amount ratio	
	Certified value [1)] [mol/mol]	Uncertainty [2)] [mol/mol]
$n(^{238}\text{Pu})/n(^{242}\text{Pu})$	0.0050666	0.0000081
$n(^{239}\text{Pu})/n(^{242}\text{Pu})$	0.0022218	0.0000026
$n(^{240}\text{Pu})/n(^{242}\text{Pu})$	0.046033	0.000046
$n(^{241}\text{Pu})/n(^{242}\text{Pu})$	0.0021930	0.0000023
$n(^{244}\text{Pu})/n(^{242}\text{Pu})$	0.00025766	0.00000059
	Mass fraction	
	Certified value [1)] [μg/g]	Uncertainty [2)] [μg/g]
^{242}Pu [3)]	86.73	0.11
Pu [3)]	91.52	0.12
	Molar mass	
	Certified value [1)] [g/mol]	Uncertainty [2)] [g/mol]
Pu [3)]	241.94417	0.00011

[1)] The certified values are traceable to the International System of Units (SI) via the values on the respective certificates of the IRMM-1027o and IRMM-290b/A3. The reference date for the plutonium amounts and isotope amount ratios is January 1, 2017.

[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)] Atomic mass: M. Wang et al: The AME2012 atomic mass evaluation (II). Tables, graphs and references.

Half-lives: Monographie BIPM-5 Table of Radionuclides, Vol.5 and ^{241}Pu half-life: Wellum R. et al. 2009 (JAAS)

PLUTONIUM IN NITRIC ACID SOLUTION		
	Isotope amount fraction (x 100)	
	Certified value [1)] [mol/mol]	Uncertainty [2)] [mol/mol]
$n(^{238}Pu)/n(Pu)$	0.47990	0.00074
$n(^{239}Pu)/n(Pu)$	0.21044	0.00023
$n(^{240}Pu)/n(Pu)$	4.3601	0.0041
$n(^{241}Pu)/n(Pu)$	0.20771	0.00021
$n(^{242}Pu)/n(Pu)$	94.7175	0.0050
$n(^{244}Pu)/n(Pu)$	0.024405	0.000056
	Isotope mass fraction (x 100)	
	Certified value [1)] [g/g]	Uncertainty [2)] [g/g]
$m(^{238}Pu)/m(Pu)$ [3)]	0.47217	0.00073
$m(^{239}Pu)/m(Pu)$ [3)]	0.20793	0.00023
$m(^{240}Pu)/m(Pu)$ [3)]	4.3260	0.0041
$m(^{241}Pu)/m(Pu)$ [3)]	0.20695	0.00021
$m(^{242}Pu)/m(Pu)$ [3)]	94.7623	0.0050
$m(^{244}Pu)/m(Pu)$ [3)]	0.024618	0.000056

[1)] The certified values are traceable to the International System of Units (SI) via the values on the respective certificates of the IRMM-1027o and IRMM-290b/A3. The reference date for the plutonium amounts and isotope amount ratios is January 1, 2017.

[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.

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1 Introduction

1.1 Background

The JRC-Geel Directorate G – Nuclear Safety and Security, Unit G.2 Standards for Nuclear Safety, Security and Safeguards (previously known as EC-JRC-IRMM) has been providing solutions of enriched plutonium isotopes for the measurements of nuclear materials using state-of-the-art analytical techniques for many years.

The International Target Values for Measurement Uncertainties in Safeguarding Nuclear Materials (ITVs) are uncertainties to be considered in judging the reliability of the measurement results of analytical techniques applied to industrial nuclear and fissile materials, which are subject to safeguards verification. In 2010, the International Atomic Energy Agency (IAEA) together with the European Safeguards Research and Development Association (ESARDA), international standardisation organisations and regional safeguards authorities, published a revised version of the ITVs [1]. The ITVs-2010 are intended to be used by nuclear plant operators and safeguards organisations as a reference of the quality of measurements necessary for nuclear material accountancy.

In this context, the most accurate analytical technique mainly used for measuring the plutonium content in nuclear materials is isotope dilution mass-spectrometry (IDMS). For this purpose, the isotope ^{242}Pu is generally used as a spike since this isotope is usually found only as a minor component in plutonium of the nuclear fuel cycle. For more than a decade now, JRC-Geel has been providing a series of enriched ^{242}Pu spikes, the so-called IRMM-049 series. However, since the last certified spike CRM of the IRMM-049 series (IRMM-049d) was approaching exhaustion, it was decided to replace it by preparing a new batch called IRMM-049e, to be certified for the first time, according to the ISO 17034:2016 [2] and ISO Guide 35:2006 [3], as part of the JRC programme to supply spike CRMs.

1.2 Choice of the material

The IRMM-049e material was prepared by a dilution of an aliquot of a ^{242}Pu stock solution in nitric acid solution and subsequent dispensing into screw-cap glass ampoules. A mass fraction of ca. 0.1 mg Pu/g solution was chosen for IRMM-049e to suit various types of plutonium measurements within the Nuclear Security and Safeguards communities.

The original stock solution was prepared by dissolving a plutonium metal of French origin with a relative mass fraction $m(^{242}\text{Pu})/m(\text{Pu})$ of ca. 94.6 % in nitric acid solution (c = 5 mol/L, p.a., Merck, Darmstadt), chemically purified (November 11, 2008) and kept under weight control for future use [4].

1.3 Design of the project

About 90 units of IRMM-049e were produced and certified according to ISO 17034:2016 [2] and ISO Guide 35:2006 [3]. The material was characterised for the amount content of ^{242}Pu by ID-TIMS with the ^{239}Pu spike CRM (IRMM-1027o), while the plutonium isotope ratios were measured by TIMS. Characterisation and homogeneity measurements were combined and carried out on 10 randomly stratified selected units out of the whole batch produced (89 units).

The certified value for the ^{242}Pu amount content was additionally confirmed using the ^{239}Pu spike CRM (IRMM-086) as part of the ongoing inter-calibration of Pu spike CRMs

[5],[6] and verified using the Pu EQRAIN-13 certified test sample (Q143, second ampoule, with a total Pu concentration of 5.3375 ± 0.0064 g/kg) during the CEA/CETAMA inter-laboratory comparison.

2 Participants

The entire project has been carried out at the European Commission, Joint Research Centre, Geel, Directorate G – Nuclear Safety & Security, Unit G.2 - Standards for Nuclear Safety, Security & Safeguards, Belgium.
This includes processing of the reference material, the chemical purification and isotope ratio measurements for the homogeneity and characterisation studies, including data evaluation and value assignment.

3 Material processing and process control

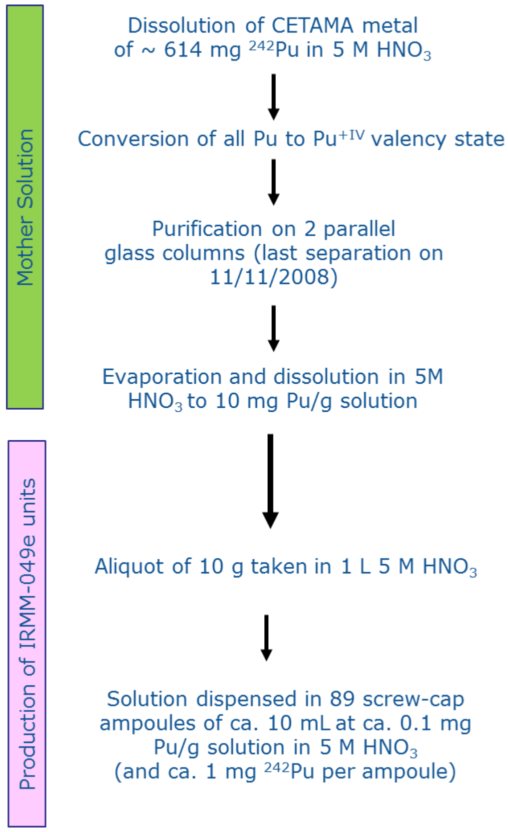
3.1 Origin and purity of starting material

The original material was prepared from a plutonium metal of French origin with a relative mass fraction $m(^{242}Pu)/m(Pu)$ of ca. 94.6 %. This metal had been dissolved in nitric acid solution (c = 5 mol/L, p.a., Merck, Darmstadt) to produce the stock solution containing about 614 mg ^{242}Pu and then purified to remove the daughter decay products [4]. This ^{242}Pu mother solution with a mass fraction of ca. 10 mg Pu/g solution was used for the preparation of the previous batch of ^{242}Pu spike CRM, the IRMM-049d, and for the plutonium fraction of the IRMM-046c (mixed $^{242}Pu/^{233}U$ spike), and as well as for the preparation of IRMM-049e (Figure 1).

3.2 Processing

The IRMM-049e units were produced by diluting an aliquot of ca. 10 g of the mother solution with 1 L nitric acid solution (c = 5 mol/L), resulting in a mass fraction of ca. 0.1 mg Pu/g solution. About 10 mL of this solution was dispensed into individual 89 screw-cap glass ampoules. The amount of total plutonium per ampoule is about 1 mg and a bit less than 1 mg for ^{242}Pu. The processing steps are shown in Figure 1.

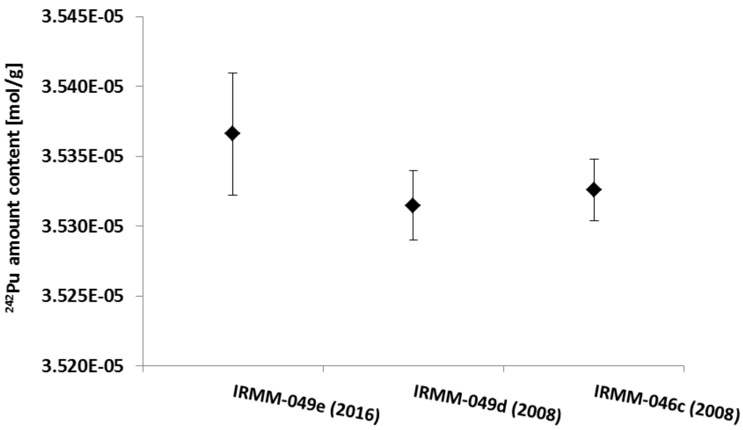
Figure 1 The processing of the IRMM-049e material



3.3 Process Control and confirmation

Estimates of the ^{242}Pu amount content of the mother solution were determined based on the weighing of the stepwise dilutions of the mother solution used to prepare the IRMM-049d, IRMM-046c and IRMM-049e units. The calculated ^{242}Pu amount contents in the mother solution from these three reference materials are in agreement within measurement uncertainties, hence confirming that there was no contamination or errors taking place during the processing of the IRMM-049e units from the initial mother solution (Figure 2).

Figure 2 Comparison of the ^{242}Pu amount contents in mol/g as estimated from the three weighing of the stepwise dilutions of the mother solution for IRMM-049e, IRMM-049d and IRMM-046c respectively (expanded uncertainties with k=2).



4 Homogeneity

A key requirement for any reference material is the equivalence between the various units. In this respect, it is relevant whether the variation between units is significant compared to the uncertainty of the certified value. In contrast to that, it is not relevant if the variation between units is significant compared to the analytical variation. Consequently, ISO 17034 [2] requires Reference Material (RM) producers to quantify the between unit variation. This aspect is covered in between-unit homogeneity studies.

The homogeneity study of IRMM-049e was combined with the characterisation assessment, i.e. the same datasets of measurement results of the ^{242}Pu amount content by IDMS and of the plutonium isotope amount ratios by TIMS were used for both assessments.

4.1 Between-unit homogeneity

The between-unit homogeneity was evaluated together with the characterisation measurements of the material to ensure that the certified values of the CRM are valid for all 89 units of the material, within the stated uncertainty.

For the between-unit homogeneity test, the number of selected units corresponds to approximately the cube root of the produced number of units of IRMM-049e, but should be at least 10. Consequently, 10 units of IRMM-049e were selected out of the 89 units using a random stratified sampling scheme covering the whole batch.

4.1.1 ^{242}Pu amount content

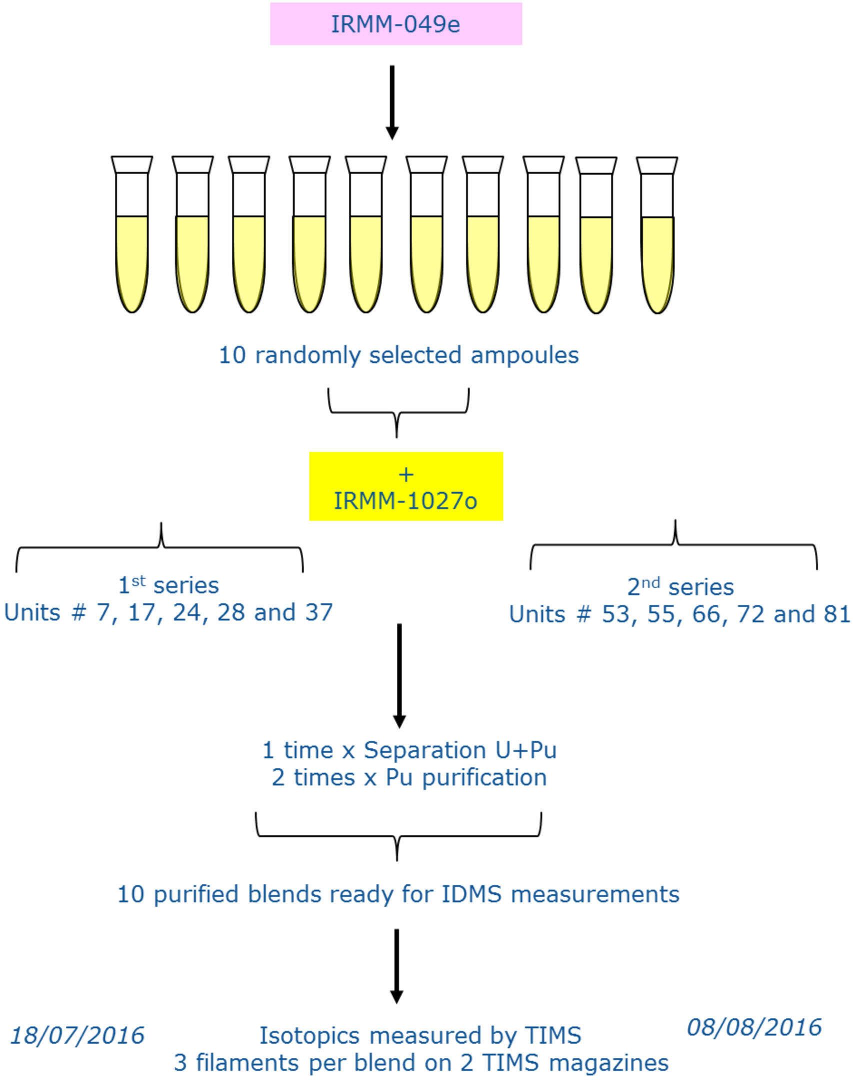
The characterisation of the plutonium amount content of IRMM-049e was established by ID-TIMS using the spike IRMM-1027o (see Annex 1) on randomly selected units using a random stratified sampling scheme (SNAP): a first series with units # 7, 17, 24, 28 and 37 and a second series with units # 53, 55, 66, 72 and 81.

Each of the 10 selected units was split in two aliquots gravimetrically: the one containing ca. 60 % of the sample was used for IDMS, the other one containing ca. 40 % of the sample was used to measure the isotopic composition or isotope abundance (IA) (Figure 3).

The IDMS blends were prepared with the Large-Sized Dried (LSD) spike IRMM-1027o (Annex 1), a ^{239}Pu spike CRM for isotope dilution [7]. This spike CRM is suitable as tracer for the IDMS measurement of IRMM-049e due to its low uncertainties of the certified values, established by a gravimetrical preparation and confirmed by IDMS. Furthermore using IRMM-1027o, the certification of IRMM-049e contributes to the on-going inter-calibration exercise of selected plutonium spikes produced by JRC-Geel on a metrological basis applying state-of-the art measurement procedures [5,6].

The blends of the IRMM-049e solution with the IRMM-1027o spike were prepared by accurate weighing using the substitution method. The following units of IRMM-1027o have been selected: 433, 444, 455, 466, 474 (first series) and 578, 599, 610, 615, 620 (second series) (see in Annex 2). Finally, 10 blends were prepared to have a $n(^{239}\text{Pu})/n(^{242}\text{Pu})$ ratio (spike to sample ratio) of ca. 0.3. The scheme for characterisation and homogeneity assessment of ^{242}Pu amount content in IRMM-049e by IDMS is shown in Figure 3.

Figure 3 Scheme of the IDMS analysis for the characterisation of the ^{242}Pu amount content in the IRMM-049e



The oxidation state of the Pu of the sample blends was adjusted to obtain Pu(IV) in 8 mol/L nitric acid and the U and Pu were separated by anion exchange method (Bio Rad, AG 1X4, 100-200 mesh) [6]. Due to the excess of uranium coming from the mixed U/Pu spike IRMM-1027o, extra purification steps were needed prior to the measurement of the Pu isotope ratios and hence determination of the Pu amount content (Figure 4). Therefore, two additional chemical separations for each blend were performed in order to obtain purified Pu fractions and thereby avoid interferences of uranium and also americium (due to ingrowth from ^{241}Pu in the starting material) with plutonium during the mass spectrometric measurements.

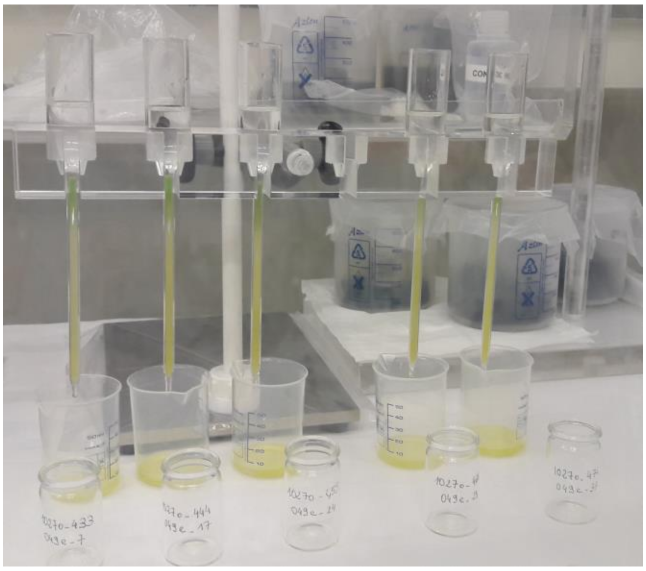
The purified fractions of Pu were prepared in nitric acid solution (c = 1 mol/L) to obtain a solution with a concentration of ca. 50 ng of Pu/µL. One µL of this solution was deposited on an evaporation rhenium filament and dried down by passing an electrical current through the filament.

Finally, two TIMS magazines were prepared with three filaments (replicates) for each blend (so 30 measurement values in total). The six remaining positions on the magazines were filled with the IRMM-290b/A3 plutonium isotopic reference material to correct for possible mass fractionation effects in the ion source. The plutonium isotope ratios $n(^{239}\text{Pu})/n(^{242}\text{Pu})$ in the blends were measured on the Triton TIMS (Thermo Fisher

Scientific, Bremen, Germany) applying a total evaporation technique [8],[9],[10]. Thus, the measurement was continued until the sample was completely exhausted in order to minimise mass fractionation effects.

The two sample magazines were measured on 18/07/2016 and 08/08/2016, respectively (see input values in Annex 2).

Figure 4 Anion exchange chromatography used to separate and purify the Pu fractions in the selected units of IRMM-049e



4.1.2 Isotopic composition of plutonium (isotope abundance ratios)

The isotopic composition of plutonium in the IRMM-049e units was established by measuring the $n(^{238}Pu)/n(^{242}Pu)$, $n(^{239}Pu)/n(^{242}Pu)$, $n(^{240}Pu)/n(^{242}Pu)$, $n(^{241}Pu)/n(^{242}Pu)$ and $n(^{244}Pu)/n(^{242}Pu)$ isotope amount ratios (IA). The measurements were carried out using the Triton TIMS on an aliquot (corresponding to ca. 40 % of the sample) of each the 10 randomly selected units (Figure 3).

Prior to mass spectrometry measurements, a chemical separation was performed using the same procedure as described in Section 4.1.1 to obtain the purified plutonium fractions. It has been proven from past experience with samples of the IRMM-049 series that have been purified twice prior to mass spectrometry measurements [6] that there was no residual ^{241}Am stemming from the decay of the ^{241}Pu in the solution.

Three replicates were measured per sample by TIMS for each series following a similar procedure as that described in Section 4.1.1 (so 30 measurement values in total for each isotope ratio). The two series of measurements were carried out on different days and the measurement results were then decay-corrected to a common date, 01/01/2017 (see in Annex 3).

4.1.3 Results for the between-unit homogeneity

For the homogeneity study, the amount content and the isotope amount ratios for the 10 units were determined. Measurements of the samples were performed in a randomised analytical sequence in order to be able to separate a potential analytical drift from a trend in the filling sequence. One data point (one value out of the 30 values) for the ^{242}Pu amount content had to be discarded because the filament broke during heating.

Since for the ^{242}Pu amount content, the 10 blends were measured on different magazines and different dates (18/07/2016 and 08/08/2016), all the values for the ^{242}Pu amount content have been decay-corrected to a reference date, arbitrarily chosen as the 01/01/2017. These measurements were hence carried out under intermediate precision conditions rather than under repeatability conditions, and in a randomised manner to be able to separate a potential analytical drift from a trend in the filling sequence.

Regression analyses were performed to evaluate potential trends in the analytical sequence as well as trends in the filling sequence. No trends could be observed in the filling sequence or the analytical sequence for the ^{242}Pu amount content and in the isotope amount ratios (see Annex 4). Note that one replicate out of the three for unit #17 was discarded since the filament broke during the measurement for the IDMS determination of the ^{242}Pu amount content.

The datasets were assessed for consistency using Grubbs outlier tests at a confidence level of 99 % on the individual results and the unit means. The homogeneity study showed no outlying unit means or trends in the filling sequence.

Quantification of between-unit inhomogeneity was accomplished by analysis of variance (ANOVA), which can separate the between-unit variation (s_{bb}) from the within-unit variation (s_{wb}). The latter is equivalent to the method repeatability if the individual samples are representative for the whole unit [11]. Therefore, the data were checked for any significant trends in the filling sequence and between the means using one way-ANOVA on the samples in analytical sequence order. The data used for the homogeneity study (measured amount contents and isotopic ratios for the 10 randomly stratified selected units) are shown in Annex 4.

Evaluation by ANOVA requires that results for each unit follow unimodal distributions with approximately the same standard deviations, and that unit means follow at least unimodal distribution. Distribution of the unit means was visually tested using histograms and normal probability plots. Minor deviations from unimodality of the individual values do not significantly affect the estimate of between-unit standard deviations.

Eventually, the results for the plutonium isotope amount ratios (see Annex 4) were not taken into account for the homogeneity study. Indeed, the reference material being a true solution and since contamination of the samples can be ruled out, there cannot be any inhomogeneity in the plutonium isotope amount ratios.

Moreover, as seen in Annex 4, no heterogeneity was found in the isotope amount ratios of all the samples. Therefore, according to ISO Guide 35:2006 [3], it is not necessary to include the contribution of the homogeneity for the plutonium isotope amount ratios in the certification.

The results of the statistical evaluations are given in Table 1.

Table 1 Results of the statistical evaluation of the homogeneity study for the ^{242}Pu amount content

Measurand	Trends*		Outliers**		Distribution	
	Analytical sequence	Filling sequence	Individual results	Unit means	Individual results	Unit means
^{242}Pu amount content [µmol/g]	no	no	none	none	normal/ unimodal	normal/ unimodal

* 95 % confidence level

** 99 % confidence level

One has to bear in mind that s_{bb} and s_{wb} are estimates of the true standard deviations and therefore subject to random fluctuations. Therefore, the mean square between groups ($MS_{between}$) can be smaller than the mean square within groups (MS_{within}), resulting in negative arguments under the square root used for the estimation of the between-unit variation, whereas the true variation cannot be lower than zero. In this case, u^*_{bb}, the maximum inhomogeneity that could be hidden by method repeatability, was calculated as described in [11]. u^*_{bb} is comparable to the limit of detection of an analytical method, yielding the maximum inhomogeneity that might be undetected by the given study setup.

Method repeatability ($s_{wb,rel}$), between–unit standard deviation ($s_{bb,rel}$) and $u^*_{bb,rel}$ were calculated as:

$$s_{wb,rel} = \frac{\sqrt{MS_{within}}}{\bar{y}}$$ **Equation 1**

$$s_{bb,rel} = \frac{\sqrt{\frac{MS_{between} - MS_{within}}{n}}}{\bar{y}}$$ **Equation 2**

$$u^*_{bb,rel} = \frac{\sqrt{\frac{MS_{within}}{n}}\sqrt[4]{\frac{2}{\nu_{MSwithin}}}}{\bar{y}}$$ **Equation 3**

MS_{within}	mean square within a unit from an ANOVA
$MS_{between}$	mean squares between-unit from an ANOVA
$\bar{y}$	mean of all results of the homogeneity study
n	number of replicates per unit
$\nu_{MSwithin}$	degrees of freedom of MS_{within}

The resulting values from the above equations were converted into relative uncertainties. The results of the evaluation of the between-unit variation are summarised in Table 2.

Table 2 Results of the homogeneity study for the ^{242}Pu amount content

Measurand	$s_{wb,rel}$ [%]	$s_{bb,rel}$ [%]	$u^*_{bb,rel}$ [%]	$u_{bb,rel}$ [1)] [%]
^{242}Pu amount content [µmol/g]	0.019	0.019	0.0062	0.019

1) standard uncertainty

For the ^{242}Pu amount content, s_{bb} was found above u^*_{bb}, the limit to detect inhomogeneity. Therefore, s_{bb} the between-unit standard deviation is used as estimate of u_{bb}.

4.2 Within-unit homogeneity and minimum sample intake

The within-unit inhomogeneity does not influence the uncertainty of the certified value when the minimum sample intake is respected, but determines the minimum size of an aliquot that is representative for the whole unit.

The within-unit homogeneity is closely correlated to the minimum sample intake. Due to this correlation, individual aliquots of a material will not contain the same amount of analyte. The minimum sample intake is the minimum amount of sample that is representative for the whole unit and thus can be used in an analysis. Sample sizes equal or above the minimum sample intake guarantee the certified value within its stated uncertainty [3].

Quantification of within-unit inhomogeneity to determine the minimum sample intake was not needed for IRMM-049e, because the material is a true solution and as such can be regarded as completely homogeneous. Therefore, no minimum sample intake should be taken into account for the analysis.

5 Stability

Stability testing is necessary to establish conditions for storage (long-term stability) as well as conditions for dispatch to the customers (short-term stability).

Temperatures up to 60 °C could be reached for regular shipment of reference materials. Therefore, stability under these conditions has to be demonstrated. The shipment of nuclear material follows the legal requirements related to radioprotection measures for transport of radioactive materials. The packing of radioactive material is divided into two parts, the packing of the inner package (Type A container) and the packing of the outer container according to regulations and respective procedures [12]. From the package material specification and the fact that the transport of radioactive material does not take longer than one week, the IRMM-049e units packed as described above are never exposed to temperatures outside the range of 4 to 60 °C. No significant change in the amount content of ^{242}Pu due to evaporation is expected to occur during shipment. The plutonium isotope amount ratios are independent on the temperature.

No dedicated long-term stability study was performed for this reference material since the general behaviour is well known from past experience with the predecessors from the IRMM-049 and IRMM-046 series. Nevertheless, this reference material will be subjected to post-certification monitoring to control its stability. Stability measurements will be done on 2 units every two years from the issuance of material certificate.

Based on the chosen approach no additional uncertainty for short-term and long-term stability will be applied.

6 Characterisation

The material characterisation is the process of determining the property value(s) of a reference material.

The material characterisation was based on the use of IDMS with IRMM-1027o as the spike CRM that had been certified by gravimetry, confirmed by ID-TIMS and externally verified by EC-JRC-Karlsruhe and the IAEA [7,13]. IDMS has the potential to be a primary ratio method of measurement.

A primary method is "a method having the highest metrological qualities, whose operation(s) can be completely described and understood and for which a complete uncertainty statement can be written in terms of SI units. A primary ratio method *measures the value of a ratio of an unknown to a standard of the same quantity; its operation must be completely described by a measurement equation* [3, 14]".

The characterisation of IRMM-049e was also verified as part of the on-going inter-calibration campaign using state-of-the art measurement procedures linking JRC spike reference materials (IRMM-086) and external plutonium quality control certified test samples (CEA/CETAMA Pu EQRAIN-13).

6.1 Method used

The characterisation assessment was combined with the homogeneity assessment, i.e. that the same dataset as the one presented in Section 4.1 was used for the determination of the uncertainty for the characterisation of the material. Hence, the method described in Section 4.1 applies to the characterisation as well. It is a validated analytical method and routinely used in Unit G.2 at JRC-Geel.

6.2 ^{242}Pu amount content and Pu isotope amount ratios and their uncertainties

The ^{242}Pu amount content in IRMM-049e was determined by IDMS [15] using the ^{239}Pu spike (IRMM-1027o), through the measurement of the isotope ratio *R(b)* = $n(^{242}\text{Pu})/n(^{239}\text{Pu})$ in a blend via the following equation:

$$c\left(^{242}Pu, x\right) = c\left(^{239}Pu, y\right) \cdot \frac{m(y)}{m(x)} \cdot \frac{R(y) - R(b)}{R(b) - R(x)} \cdot R(x)$$ **Equation 4**

where

c(²⁴²Pu, x): amount content of ^{242}Pu in the IRMM-049e

c(²³⁹Pu, y): amount content of ^{239}Pu in the spike IRMM-1027o

m(x): mass of the IRMM-049e

m(y): mass of the spike

R(b): isotope amount ratio $n(^{242}\text{Pu})/n(^{239}\text{Pu})$ of the blend

R(x): isotope amount ratio $n(^{242}\text{Pu})/n(^{239}\text{Pu})$ of the unspiked IRMM-049e

R(y): isotope amount ratio $n(^{242}\text{Pu})/n(^{239}\text{Pu})$ of the spike IRMM-1027o

The masses of the ten selected IRMM-049e (m_x) and IRMM-1027o (m_y) units for IDMS, as shown in Annex 2, were determined by substitution weighing. In substitution weighing, the mass of a sample is determined through a series of mass determinations of an unknown (U) and a reference weight (S). The so called "SUUS" method was applied. The uncertainty contributions in substitution weighing of the samples are the uncertainties associated with the calibrated weights (certificate), air buoyancy correction and the variability of the balance used in "SUUS" method.

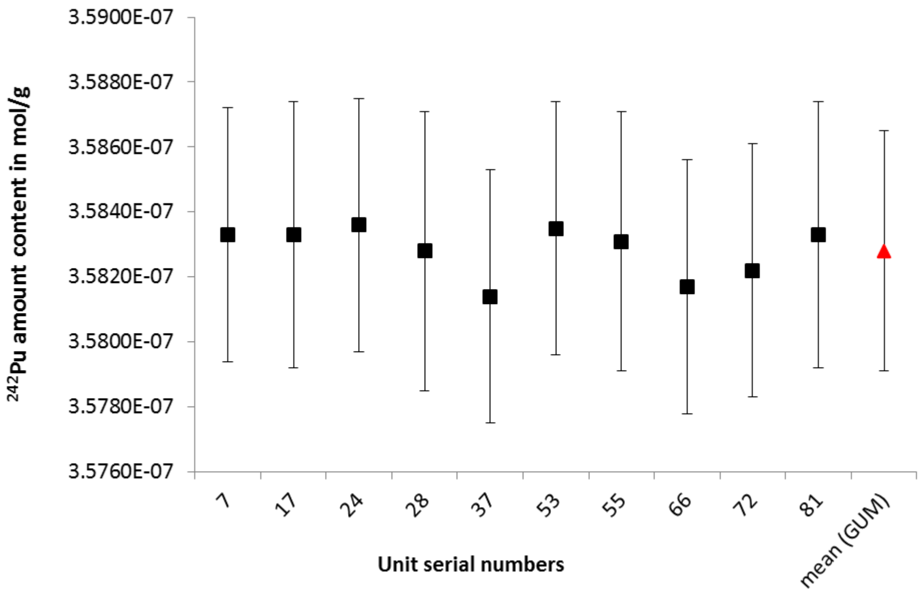
The results of these IDMS measurements for the ^{242}Pu amount content are given in Figure 5.

The certified value for the ^{242}Pu amount content was calculated as the mean value of the ten certification measurement results obtained by IDMS using the IRMM-1027o LSD spike. A consistency check was carried out according to [16] for the evaluation of the uncertainties of the ten individual sample measurements for the amount content of ^{242}Pu. The detailed IDMS equations and calculations using the software GUM Workbench [17] can be found in Annex 2.

The certified values for the isotope amount ratios result from the mean of the measured values for the ten randomly stratified selected units measured by mass spectrometry TIMS. The detailed equations and calculations using the software GUM Workbench [17] can be found in Annex 3.

For all the measurands, full uncertainty budgets were established in accordance with the 'Guide to the Expression of Uncertainty in Measurement' [18].

Figure 5 The ^{242}Pu amount content in mol/g in the 10 units of IRMM-049e measured by IDMS using IRMM-1027o as spike CRM (with associated expanded uncertainties k = 2)



The results of the evaluation of the characterisation study are summarised in Table 3.

Table 3 Standard uncertainties of characterisation for IRMM-049e

Measurand	u_{char} [1)]	$u_{char, rel}$[%] [1)]
^{242}Pu amount content [µmol/g]	0.000214	0.060
$n(^{238}Pu)/n(^{242}Pu)$ [mol/mol]	$4.03 \cdot 10^{-6}$	0.080
$n(^{239}Pu)/n(^{242}Pu)$ [mol/mol]	$1.28 \cdot 10^{-6}$	0.058
$n(^{240}Pu)/n(^{242}Pu)$ [mol/mol]	$23.0 \cdot 10^{-6}$	0.050
$n(^{241}Pu)/n(^{242}Pu)$ [mol/mol]	$1.13 \cdot 10^{-6}$	0.052
$n(^{244}Pu)/n(^{242}Pu)$ [mol/mol]	$0.29 \cdot 10^{-6}$	0.11

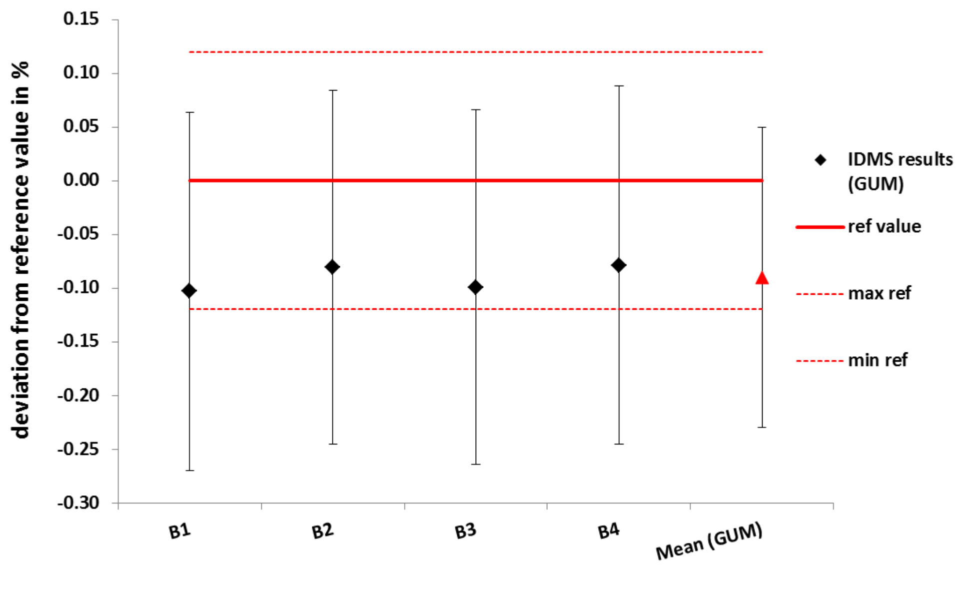
1) Standard uncertainty

6.3 Confirmation measurements

The ^{242}Pu amount content in IRMM-049e was first verified in the framework of the plutonium inter-laboratory comparison, EQRAIN-13, organised by the CEA/CETAMA [19]. The ampoule Q143 of EQRAIN-13, with a total Pu concentration of (5.3375 ± 0.0064) g/kg, was gravimetrically mixed with IRMM-049e into four blends in order to determine the total Pu mass content (in g/g) in the EQRAIN-13 ampoule using the ^{242}Pu in IRMM-049e as spike for ID-TIMS. The IDMS results from these measurements were then compared to the reference value provided by CEA/CETAMA. Full uncertainty budgets were established in accordance with the 'Guide to the Expression of Uncertainty in Measurement' [18]. Figure 6 shows the IDMS results for the total Pu mass content in the four blends of EQRAIN-13 (Q143) with IRMM-049e as relative deviation from the EQRAIN-13 reference value. Although there is an observed systematic slight deviation (≤ 0.1 %) from the given reference value, the IDMS results agree within the uncertainties with the reference value, confirming the value for the ^{242}Pu amount content and amount ratio $n(^{239}\text{Pu})/n(^{242}\text{Pu})$ from the characterisation of IRMM-049e with IRMM-1027o. Furthermore, the normalized deviation applied on IDMS results on Pu EQRAIN-13 is 0.30 % [19] and therefore encompasses this observed deviation of 0.1 %.

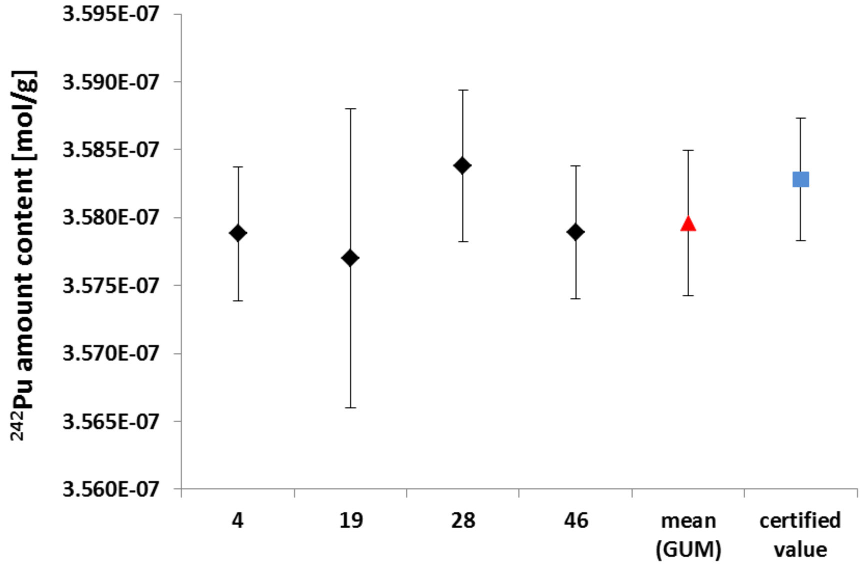
Moreover, JRC-Geel's laboratory performance in EQRAIN-13 was satisfactory (see lab 19, by IDMS technique in Annex 5), as expressed by means of *z*- and *ζ*- scores to the reference value and the standard deviation for proficiency assessment derived from participants' results reported with 7 different analytical techniques. This, and the fact that the required relative standard uncertainty for plutonium element concentration measurements (DA) carried out under glove box conditions using small size spikes in the ITV-2010 [1] is 0.28 % ($k = 1$), provides an external verification for the characterisation of IRMM-049e and demonstrates that this spike CRM is fit-for-purpose.

Figure 6 IDMS results for the total Pu mass fraction (in g/g) in EQRAIN-13 (second ampoule, Q143) determined using IRMM-049e



Moreover, the reference material values to be certified were confirmed by ID-TIMS using another CRM, the ^{239}Pu spike, IRMM-086. In September 2016, four units of IRMM-086 and two units of IRMM-049e were randomly selected and gravimetrically mixed into four blends for Isotope Dilution. After the necessary chemical separations/purifications, the four blends were loaded on a TIMS magazine, with three filaments (replicates) for each blend and measured using the total evaporation (TE) technique on the TRITON TIMS. Figure 7 shows the IDMS results for the ^{242}Pu amount content in mol/g in the four blends of IRMM-049e using IRMM-086 as spike CRM, their mean and the certified value of IRMM-049e using IRMM-1027o as spike CRM, see also Table 4). This figure shows that the results with IRMM-086 overlap and agree within the uncertainties with the final certified value of the ^{242}Pu amount content of IRMM-049e.

Figure 7 IDMS results for the ^{242}Pu amount content [mol/g] in IRMM-049e determined using IRMM-086 as spike CRM



7 Value assignment

Certified values are values that fulfil the highest standards of accuracy. Procedures at JRC-Geel require generally pooling of not less than 6 datasets to assign certified values. Full uncertainty budgets in accordance with the 'Guide to the Expression of Uncertainty in Measurement' [18] were established.

Finally, the assigned uncertainty consists of uncertainties related to characterisation u_{char} (Section 6.2) and potential between-unit inhomogeneity, u_{bb} (Section 4.1)

These different contributions were combined to estimate the expanded uncertainty of the certified value (U_{CRM}) with a coverage factor k as:

$$U_{CRM} = k \cdot \sqrt{u_{char}^2 + u_{bb}^2} \qquad \textbf{Equation 5}$$

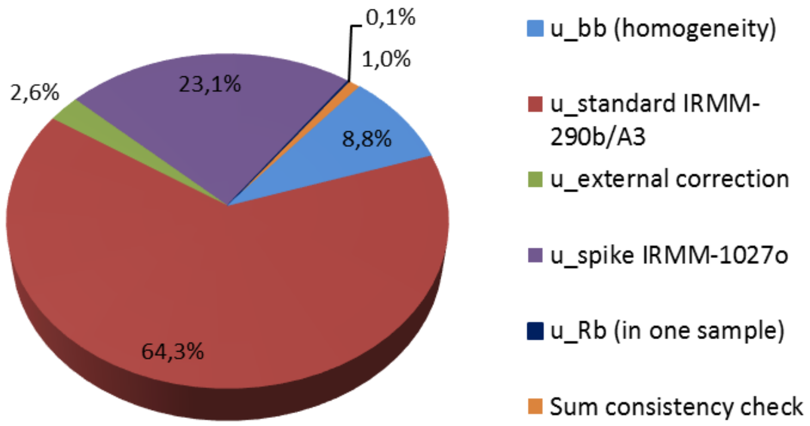
- u_{char} was estimated as described in Section 6.2
- u_{bb} was estimated as described in Section 4.1

Because of the sufficient numbers of the degrees of freedom of the different uncertainty contributions, a coverage factor k of 2 was applied, to obtain the expanded uncertainties. The certified values and their uncertainties are summarised in Table 4 and Table 5.

The major component to the expanded uncertainties for the ^{242}Pu amount content and isotope amount ratios comes from the standard IRMM-290b/A3 used for the mass bias correction during the TIMS measurements, and for the ^{242}Pu amount content, it stems as well from the ^{239}Pu spike IRMM-1027o used for the Isotope Dilution. Figure 8 and Figure 9 show the relative contributions of the main uncertainty components to the overall uncertainties of the certified values for the ^{242}Pu amount content and the $^{239}Pu/^{242}Pu$ amount ratio in IRMM-049e respectively.

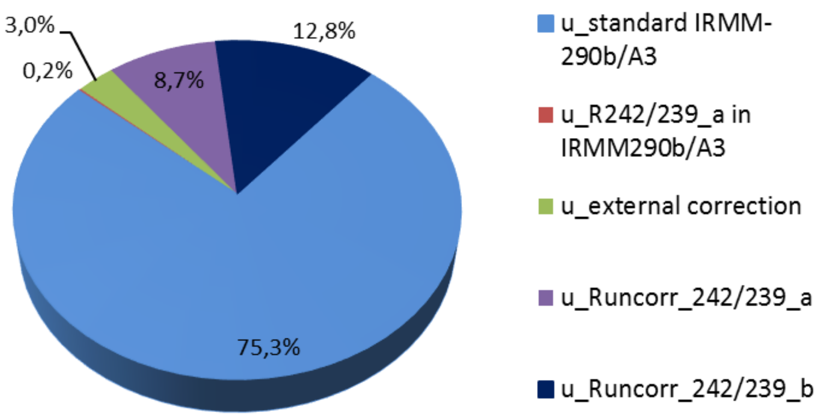
Therefore for the other certified values that are directly derived from the ^{242}Pu amount content such as the mass fractions, or from the amount ratios as the abundances in mole and mass fractions, the major component to the expanded uncertainties comes as well from the standard IRMM-290b/A3 (see as well budgets as reported in Annex 2 and Annex 3).

Figure 8 Uncertainty budget of the major components (as standard uncertainties) on the certified ^{242}Pu amount content [µmol/g] in IRMM-049e as on 01/01/2017 and calculated in GUM Workbench



Where u_bb represents the uncertainty coming from the homogeneity assessment (i.e. 0.019 %, *k* = 1, of certified value), u_standard IRMM 290b/A3 is the uncertainty of the IRMM-290/A3 standard (of 0.1 %, *k* = 2), u_external correction is 0.01 %, *k* = 1, u_spike IRMM-1027o corresponds to the uncertainty of the spike (of 0.06 %, *k* = 2), u_Rb the uncertainty of the ratio in the blend as measured by ID-TIMS and the sum of the consistency check correction represents the uncertainty of the delta values [16], which is here of $140 \cdot 10^{-12}$ (see also Annex 2).

Figure 9 Uncertainty budget of the major components (as standard uncertainties) on the certified $^{239}Pu/^{242}Pu$ amount ratio [mol/mol] in IRMM-049e as on 01/01/2017 and calculated in GUM Workbench



Where u_Runcorr_242/239_a or b are the uncertainties of the measured $^{242}Pu/^{239}Pu$ on the TIMS turrets (F) and (E) respectively (see as well Annex 3).

7.1 Certified values and their uncertainties

Table 4 Certified ^{242}Pu and total Pu amount content values and the uncertainties in IRMM-049e

	Certified value [µmol/g]	U_{CRM} 1) [µmol/g]	U_{CRM} 1) [%]
^{242}Pu amount content	0.35828	0.00045	0.13
Pu amount content	0.37826	0.00047	0.13

1) Expanded (k = 2) uncertainty

Table 5 Certified isotope amount ratio values and their uncertainties in IRMM-049e

	Certified value [mol/mol]	U_{CRM} 1) 2) [mol/mol]	$U_{CRM, rel}$ 1) 2) [%]
$n(^{238}Pu)/n(^{242}Pu)$	0.0050666	0.0000081	0.16
$n(^{239}Pu)/n(^{242}Pu)$	0.0022218	0.0000026	0.12
$n(^{240}Pu)/n(^{242}Pu)$	0.046033	0.000046	0.10
$n(^{241}Pu)/n(^{242}Pu)$	0.0021930	0.0000023	0.10
$n(^{244}Pu)/n(^{242}Pu)$	0.00025766	0.00000059	0.23

1) Expanded (k = 2) uncertainty

2) Note that for the isotope amount ratios the contribution of the homogeneity has not been taken into account

Using the certified values listed above for the ^{242}Pu amount contents and the isotope amount ratios, the uncertainty budgets for the mass fractions, and the abundance mole and mass fractions of Pu isotopes were set up as for the certified values, according to the 'Guide to the Expression of Uncertainty in Measurement' [18], and are listed in Table 6, Table 7 and Table 8 respectively.

Table 6 Certified mass fractions and their uncertainties in IRMM-049e

	Certified value 1) [µg/g]	U_{CRM} 1) [µg/g]	$U_{CRM, rel}$ 1) [%]
^{242}Pu	86.73	0.11	0.13
Pu	91.52	0.12	0.13

1) Expanded (k = 2) uncertainty

The molar mass (g/mol) of plutonium was determined as well using GUM Workbench (see Annex 3) and its certified value is (241.94417 ± 0.00011) g/mol, as reported as well in the certificate of the IRMM-049e.

Table 7 Certified abundances as mole fractions and their uncertainties for IRMM-049e

Mole fraction (x100)	Certified value [mol/mol]	U_{CRM} 1) [mol/mol]	$U_{CRM, rel}$ 1) [%]
$n(^{238}Pu)/n(Pu)$	0.47990	0.00074	0.16
$n(^{239}Pu)/n(Pu)$	0.21044	0.00023	0.11
$n(^{240}Pu)/n(Pu)$	4.3601	0.0041	0.094
$n(^{241}Pu)/n(Pu)$	0.20771	0.00021	0.10
$n(^{242}Pu)/n(Pu)$	94.7175	0.0050	0.0053
$n(^{244}Pu)/n(Pu)$	0.024405	0.000056	0.23

1) Expanded (k = 2) uncertainty

Table 8 Certified abundances as mass fractions and their uncertainties for IRMM-049e

Mass fraction (x100)	Certified value [g/g]	U_{CRM} 1) [g/g]	$U_{CRM, rel}$ 1) [%]
$m(^{238}Pu)/m(Pu)$	0.47217	0.00073	0.16
$m(^{239}Pu)/m(Pu)$	0.20793	0.00023	0.11
$m(^{240}Pu)/m(Pu)$	4.3260	0.0041	0.10
$m(^{241}Pu)/m(Pu)$	0.20695	0.00021	0.10
$m(^{242}Pu)/m(Pu)$	94.7623	0.0050	0.0052
$m(^{244}Pu)/m(Pu)$	0.024618	0.000056	0.23

1) Expanded (k = 2) uncertainty

8 Metrological traceability and commutability

8.1 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 Pu amount content value is traceable to the certified values of the certificate of the IRMM-1027o LSD spike.

The Pu amount isotope ratio values are traceable to the certified values on the IRMM-290b/A3 certificate.

Material used as second "calibrant" in the overall metrology pyramid.

8.2 Commutability

Many measurement procedures include one or more steps, which select specific -or specific groups of- 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 (methods) 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 [20] 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 and is for use as a spike isotopic reference material for the IDMS analysis of a wide range of nuclear samples, mainly by the nuclear and safeguards communities.

9 Instructions for use

9.1 Safety information

The usual laboratory safety measures apply for the handling of radioactive/nuclear material.

9.2 Storage conditions

The vials should be stored at + 18 °C ± 5 °C.

The user is reminded to close the vial(s) immediately after taking the sample.

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

9.3 Preparation and use of the material

This material is ready for use after ampoule opening. Users are cautioned that once the ampoule is opened, the amount content of plutonium may be affected by evaporation losses.

The IRMM-049e is intended for use as a spike isotopic reference material for IDMS analysis of a wide range of nuclear samples.

9.4 Minimum sample intake

Because the IRMM-049e material is a true solution and as such, can be regarded as completely homogeneous, there is no minimum sample intake to be taken into account for the analysis.

9.5 Use of the certified value

The main purpose of this material is to assess method performance, i.e. for checking accuracy of analytical results and calibration. As any reference material, it can also be used for control charts or validation studies.

<u>Use as a spike isotopic reference material</u>

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

<u>Comparing an analytical result with the certified value</u>

A result can be considered as 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 [21]).

For assessing the method performance, the measured values of the CRMs are compared with the certified values. The procedure is described here in brief:

- Calculate the absolute difference between mean measured value and the certified value (Δ_{meas})
- Combine measurement uncertainty (u_{meas}) with the uncertainty of the certified value (u_{CRM}): $u_\Delta = \sqrt{u_{meas}^2 + u_{CRM}^2}$
- 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 %
- If $\Delta_{meas} \le U_\Delta$ no significant difference between the measurement result and the certified value, at a confidence level of about 95 % exists

<u>Use for quality control purpose</u>

The certified values can be used for quality control charts. Different CRM-units will give the same result, because inhomogeneity was included in the uncertainties of the certified values.

10 Conclusions

A new highly enriched ^{242}Pu isotopic reference material IRMM-049e has been prepared and certified for the ^{242}Pu plutonium isotope amount content and isotopic composition. The IRMM-049e isotopic reference material is supplied in a glass ampoule with screw cap seals containing about 10 mL of 5 mol/L nitric acid with plutonium. The content of each vial is approximately 1 mg ^{242}Pu. The certified values for amount content have been established by isotope dilution mass spectrometry and confirmed as part of the Pu spike inter-calibration campaign and verified via participation in the CEA/CETAMA Pu EQRAIN-13 inter-laboratory comparison. The IRMM-049 series are Isotopic Reference Materials applied in nuclear fuel cycle measurements and are part of a systematic JRC-Geel programme to supply spike isotopic reference materials of various isotopes at different concentrations.

References

[1] International Target Values 2010 for Measurement Uncertainties in Safeguarding Nuclear Materials, IAEA-STR-368, Vienna, November 2010.

[2] ISO 17034:2016, General requirements for the competence of reference materials producers, International Organization for Standardization, Geneva, Switzerland, 2016.

[3] ISO Guide 35, Reference materials – General and statistical principles for certification, International Organization for Standardization, Geneva, Switzerland, 2006.

[4] Jakopič, R., Eykens, R., Kehoe, F., Kühn, H., Richter, S., Verbruggen, A., et al. (2011). *Preparation and certification of the Isotopic Reference Material IRMM-049d.* Report EUR 24915 EN.

[5] R. Jakopič, J. Bauwens, S. Richter, M. Sturm, A. Verbruggen, R. Wellum, R. Eykens, F. Kehoe, H. Kühn, Y. Aregbe (2011). Preparation and development of new Pu spike isotopic reference materials at IRMM, ESARDA Bulletin, No. 46.

[6] R. Jakopič, A. Verbruggen, R. Eykens, F. Kehoe, H. Kühn, Y. Kushigeta, U. Jacobsson, J. Bauwens, S. Richter, R. Wellum, Y. Aregbe (2010). An inter-calibration campaign using various selected Pu spike isotopic reference materials, J. Radioanal. Nucl. Chem., 286, 449-454.

[7] Jakopič, R., Bauwens, J., Buják, R., Eykens, R., Henessy, C., Kehoe, F., et al. (2013). *Preparation and certification of Large-Sized Dried (LSD) Spike IRMM-1027o.* Report EUR 25857 EN.

[8] Fiedler, R., Donohue, D., Grabmueller, G., & Kurosawa, A. (1994). Report on preliminary experience with total evaporation measurements in thermal ionization mass spectrometry. *International Journal of Mass Spectrometry and Ion Processes, 132*, 207-215.

[9] Richter, S., Kuehn, H., Aregbe, A., Hedberg, M., Horta-Domenech, J., Mayer, K., et al. (2011). Improvements in routine uranium isotope ratio measurements using the modified total evaporation method for multi-collector thermal ionization mass spectrometry. *J. Anal. At. Spectrom., 26*, 550.

[10] ASTM Standard C1672-17 Standard Test Method for Determination of Uranium or Plutonium Isotopic Composition or Concentration by the Total Evaporation Method using a Thermal Ionization Mass Spectrometer. DOI: 10.1520/C1672-17.

[11] Linsinger, T. P., Pauwels, J., van der Veen, A., Schimmel, H., and Lamberty, A. (2001). Homogeneity and stability of reference materials. Accreditation and Quality Assurance, 6, 20-25.

[12] IAEA Regulations for the Safe Transport of Radioactive Material, 1996 Edition, "Requirements", IAEA Safety Standards Series No. TS-R-1 (ST-1, Revised), IAEA, Vienna, 2000

[13] R. Jakopič, Y. Aregbe, S. Richter, E. Zuleger, S. Mialle, S. D. Balsley, U. Repinc, J. Hiess (2017). Verification measurements of the IRMM-1027 and the IAEA large-sized dried (LSD) spikes J Radioanal Nucl Chem 311:1781–1791.

[14] Milton, M. J., & Quinn, T. J. (2001). Primary methods for the measurement. Metrologia, 38, 289-296.

[15] De Bievre, P., & Peiser, H. S. (1997). Basic equations and uncertainties in isotope-dilution mass spectrometry for traceability to SI of values obtained by this primary method. Fresenius. Journal of Analytical Chemistry (359), 523-525.

[16] Kessel, R., Berglund, M., & Wellum, R. (2008). Application of consistency checking to evaluation of uncertainty in multiple replicate measurements. *Accred Qual Assur*. DOI 10.1007/s00769-008-0382-x

[17] Programme GUM work bench 2.4, Metrodata GmbH, (2009). Weil am Rhein, Germany.

[18] ISO/IEC Guide 98-3, Guide to the Expression of Uncertainty in Measuremen;t, (GUM 1995), International Organization for Standardization, Geneva, Switzerland, 2008

[19] Report (in French) CEA/CETAMA Circuit EQRAIN Plutonium n°13 – Essai d'aptitude par comparaison interlaboratoires- Synthese du programme EQRAIN "plutonium" N13 (2014-2016). CEA/DEN/MAR/DMRC/CETAMA/NT/2017/01 ind. A.

[20] Vesper, H., Emons, H., Gnezda, M., Jain, C. P., Miller, W. G., Rej, R., Schumann, G., Tate, J., Thienpont, L., Vaks, J. E. (2010). Characterization and Qualification of Commutable Reference Materials for Laboratory Medicine; Approved Guideline, CLSI document C53-A, Clinical and Laboratory Standards Institute, Wayne, PA, USA.

[21] Linsinger, T.P.J. (2010) ERM Application Note 1: Comparison of a measurement result with the certified value, www.erm-crm.org (last accessed on 06/06/2017).

List of abbreviations and definitions

ANOVA	Analysis of variance
c	Amount of substance concentration
CEA	Le Commissariat à l'énergie atomique et aux énergies alternatives, la Direction de l'énergie nucléaire (Alternative Energies and Atomic Energy Commission)
CETAMA	Commission d'Etablissement des Méthodes d'Analyse
CLSI	Clinical and Laboratory Standards Institute
CRM	Certified reference material
DA	Destructive analysis
EC	European Commission
ESARDA	European Safeguards Research and Development Association
EQRAIN	Quality Assessment of Analysis Results in Nuclear Industry
GUM	Guide to the Expression of Uncertainty in Measurement
IAEA	International Atomic Energy Agency
IDMS	Isotope dilution mass spectrometry
ID-TIMS	Isotope dilution thermal ionisation mass spectrometry
ISO	International Organization for Standardization
ITVs	International Target Values
JRC	Joint Research Centre of the European Commission
k	Coverage factor
LSD	Large-Sized Dried
m	Mass
M	Molar mass
MS	Mass spectrometry
$MS_{between}$	Mean of squares between-unit from an ANOVA
MS_{within}	Mean of squares within-unit from an ANOVA
n	Amount of substance
N	Mean number of replicates per unit
p.a.	Pro analysis
rel	Index denoting relative figures (uncertainties etc.)
RM	Reference material
s	Standard deviation
s_{bb}	Between-unit standard deviation; an additional index "rel" is added when appropriate
SI	International System of Units
s_{wb}	Within-unit standard deviation; an additional index "rel" is added

	when appropriate
TIMS	Thermal Ionisation Mass Spectrometry
u	Standard uncertainty
U	Expanded uncertainty
u^*_{bb}	Standard uncertainty related to a maximum between-unit inhomogeneity that could be hidden by method repeatability; an additional index "rel" is added as appropriate
u_{bb}	Standard uncertainty related to a possible between-unit inhomogeneity; an additional index "rel" is added as appropriate
u_{char}	Standard uncertainty of the material characterisation; an additional index "rel" is added as appropriate
u_{CRM}	Combined standard uncertainty of the certified value; an additional index "rel" is added as appropriate
U_{CRM}	Expanded uncertainty of the certified value; an additional index "rel" is added as appropriate
u_{Δ}	Combined standard uncertainty of measurement result and certified value
u_{meas}	Standard measurement uncertainty
$\overline{y}$	Arithmetic mean
$\nu_{MSwithin}$	Degrees of freedom of MS_{within}

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Annexes

Annex 1. Certificate of the IRMM-1027o

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 – 1027o

CERTIFICATE OF ANALYSIS

Uranium and Plutonium in Cellulose Acetate Butyrate (CAB)

	Isotope amount ratio	
	Certified value [1)] [mol/mol]	Uncertainty [2)] [mol/mol]
$n(^{235}U)/n(^{238}U)$	0.24242	0.00005
$n(^{240}Pu)/n(^{239}Pu)$	0.022422	0.000006
$n(^{241}Pu)/n(^{239}Pu)$	0.0001793	0.0000024
$n(^{242}Pu)/n(^{239}Pu)$	0.0000757	0.0000008

The certified masses and uncertainties of ^{235}U, ^{238}U and ^{239}Pu per unit are listed in Annex 1 on pages 3 to 4 of this certificate.

1) The certified values are traceable to the values on the respective metal certificates (EC NRM 101, NBL CRM-116 and CETAMA MP2). The reference date for the plutonium and uranium isotope amount ratios is November 1, 2012.

2) The certified 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.

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

Geel, May 2013

Last revision January 2017

Signed: 18/01/17

Dr. Willy Mondelaers
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

All following pages are an integral part of the certificate.

DESCRIPTION OF THE MATERIAL

The IRMM-1027o series of Large-Sized Dried (LSD) spikes consists of 1215 units, each containing approximately 50 mg of uranium and 1.8 mg of Pu in dried form. Units were prepared by aliquoting of about 2.5 g of a gravimetrically prepared nitrate solution of uranium (EC NRM 101 and NBL CRM-116) and plutonium (CETAMA MP2) into individual vials. The solution in each vial was dried down, re-dissolved in cellulose acetate butyrate (CAB) and dried again to produce a stable layer at the bottom of the vial. Each unit contains a unique quantity of uranium and plutonium and is assigned a serial number for identification and reference.

ANALYTICAL METHODS USED FOR CERTIFICATION

The certified values are based on the gravimetric preparation of the mother solution taking into account the isotopic composition and the purity of the starting materials, their masses and the mass of the solution. The confirmatory measurements were performed by isotope dilution thermal ionisation mass spectrometry (ID-TIMS) and thermal ionisation mass spectrometry (TIMS).

All the work related to the preparation and certification of this CRM has been performed at the European Commission, Joint Research Centre, Directorate G – Nuclear Safety and Security, G.2 – Standards for Nuclear Safety, Security and Safeguards Unit in Geel, Belgium.

SAFETY INFORMATION

The IRMM-1027o series contains radioactive material. The vials 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 Certified Reference Material (CRM) is used as a calibrant in the analysis of plutonium and uranium materials by isotope dilution mass spectrometry. The spike has to be dissolved in the appropríoate amount of acid (e.g. nitric acid with an amount of substance concentration $c = 5$ mol L^{-1}) or sample solution to ensure the isotopic equilibrium between the spike and the sample. The whole amount of sample per unit should be used for analysis.

STORAGE

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

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NOTE

A technical report on the production of IRMM-1027o is available on the internet. A paper copy can be obtained from JRC Directorate G – Nuclear Safety and Security, G.2 – Standards for Nuclear Safety, Security and Safeguards Unit in Geel, Belgium on request.

Annex 1: The certified masses of ^{238}U, ^{235}U and ^{239}Pu per unit of IRMM-1027o.

	^{238}U		^{235}U		^{239}Pu	
Vial No	Mass [1)] [mg]	Uncertainty [2)] [mg]	Mass [1)] [mg]	Uncertainty [2)] [mg]	Mass [1)] [mg]	Uncertainty [2)] [mg]
0433	43.387	0.025	10.385	0.005	1.7694	0.0009
0434	43.413	0.025	10.391	0.005	1.7704	0.0009
0435	43.464	0.025	10.403	0.005	1.7725	0.0009
0436	43.298	0.025	10.364	0.005	1.7658	0.0009
0437	43.492	0.025	10.410	0.005	1.7737	0.0009
0438	43.301	0.025	10.364	0.005	1.7659	0.0009
0439	43.483	0.025	10.408	0.005	1.7733	0.0009
0440	43.308	0.025	10.366	0.005	1.7662	0.0009
0441	43.372	0.025	10.381	0.005	1.7688	0.0009
0442	43.420	0.025	10.393	0.005	1.7707	0.0009
0443	43.435	0.025	10.396	0.005	1.7714	0.0009
0444	43.499	0.025	10.412	0.005	1.7739	0.0009
0445	43.284	0.025	10.360	0.005	1.7652	0.0009
0446	43.399	0.025	10.388	0.005	1.7699	0.0009
0447	43.415	0.025	10.392	0.005	1.7705	0.0009
0450	43.480	0.025	10.407	0.005	1.7732	0.0009
0452	43.449	0.025	10.400	0.005	1.7719	0.0009
0454	43.377	0.025	10.383	0.005	1.7690	0.0009
0455	43.404	0.025	10.389	0.005	1.7701	0.0009
0456	43.415	0.025	10.392	0.005	1.7705	0.0009
0457	43.367	0.025	10.380	0.005	1.7686	0.0009
0458	43.397	0.025	10.387	0.005	1.7698	0.0009
0459	43.353	0.025	10.377	0.005	1.7680	0.0009
0460	42.636	0.025	10.205	0.005	1.7388	0.0009
0461	43.367	0.025	10.380	0.005	1.7686	0.0009
0462	43.355	0.025	10.377	0.005	1.7681	0.0009
0463	43.451	0.025	10.400	0.005	1.7720	0.0009
0464	43.351	0.025	10.376	0.005	1.7679	0.0009
0465	43.516	0.025	10.416	0.005	1.7746	0.0009
0466	43.264	0.025	10.355	0.005	1.7644	0.0009
0467	42.605	0.025	10.198	0.005	1.7375	0.0009
0468	43.346	0.025	10.375	0.005	1.7677	0.0009
0469	43.391	0.025	10.386	0.005	1.7695	0.0009
0470	43.385	0.025	10.385	0.005	1.7693	0.0009
0471	43.428	0.025	10.395	0.005	1.7711	0.0009
0473	43.391	0.025	10.386	0.005	1.7695	0.0009
0474	43.370	0.025	10.381	0.005	1.7687	0.0009
0475	42.624	0.025	10.202	0.005	1.7383	0.0009
0476	43.325	0.025	10.370	0.005	1.7669	0.0009
0477	43.360	0.025	10.378	0.005	1.7683	0.0009
0478	42.638	0.025	10.206	0.005	1.7388	0.0009
0479	43.406	0.025	10.390	0.005	1.7702	0.0009
0480	43.262	0.025	10.355	0.005	1.7643	0.0009
0577	42.661	0.025	10.211	0.005	1.7398	0.0009
0578	43.262	0.025	10.355	0.005	1.7643	0.0009
0579	43.389	0.025	10.385	0.005	1.7695	0.0009
0580	43.379	0.025	10.383	0.005	1.7691	0.0009
0581	43.358	0.025	10.378	0.005	1.7682	0.0009
0582	43.394	0.025	10.387	0.005	1.7697	0.0009
0583	43.298	0.025	10.364	0.005	1.7658	0.0009

Annex 1: The certified masses of ^{238}U, ^{235}U and ^{239}Pu per unit of IRMM-1027o.

	^{238}U		^{235}U		^{239}Pu	
Vial No	Mass [1] [mg]	Uncertainty [2] [mg]	Mass [1] [mg]	Uncertainty [2] [mg]	Mass [1] [mg]	Uncertainty [2] [mg]
0584	43.327	0.025	10.371	0.005	1.7670	0.0009
0585	43.413	0.025	10.391	0.005	1.7705	0.0009
0586	43.360	0.025	10.378	0.005	1.7683	0.0009
0587	43.286	0.025	10.361	0.005	1.7653	0.0009
0588	42.609	0.025	10.199	0.005	1.7377	0.0009
0589	43.312	0.025	10.367	0.005	1.7663	0.0009
0590	43.276	0.025	10.358	0.005	1.7649	0.0009
0591	43.413	0.025	10.391	0.005	1.7705	0.0009
0592	43.461	0.025	10.403	0.005	1.7724	0.0009
0593	43.255	0.025	10.353	0.005	1.7640	0.0009
0594	43.432	0.025	10.396	0.005	1.7712	0.0009
0595	43.286	0.025	10.361	0.005	1.7653	0.0009
0597	43.269	0.025	10.357	0.005	1.7646	0.0009
0598	43.333	0.025	10.372	0.005	1.7672	0.0009
0599	43.363	0.025	10.379	0.005	1.7684	0.0009
0600	42.635	0.025	10.205	0.005	1.7387	0.0009
0601	43.264	0.025	10.356	0.005	1.7644	0.0009
0602	43.355	0.025	10.377	0.005	1.7681	0.0009
0603	43.420	0.025	10.393	0.005	1.7707	0.0009
0604	43.312	0.025	10.367	0.005	1.7663	0.0009
0605	43.384	0.025	10.384	0.005	1.7693	0.0009
0606	43.381	0.025	10.383	0.005	1.7691	0.0009
0607	43.381	0.025	10.383	0.005	1.7691	0.0009
0608	43.398	0.025	10.388	0.005	1.7698	0.0009
0610	43.415	0.025	10.392	0.005	1.7705	0.0009
0611	43.519	0.025	10.417	0.005	1.7748	0.0009
0612	43.321	0.025	10.369	0.005	1.7667	0.0009
0613	43.298	0.025	10.364	0.005	1.7658	0.0009
0614	43.312	0.025	10.367	0.005	1.7663	0.0009
0615	43.374	0.025	10.382	0.005	1.7688	0.0009
0616	43.405	0.025	10.389	0.005	1.7701	0.0009
0617	43.422	0.025	10.393	0.005	1.7708	0.0009
0618	43.245	0.025	10.351	0.005	1.7636	0.0009
0619	42.635	0.025	10.205	0.005	1.7387	0.0009
0620	43.321	0.025	10.369	0.005	1.7667	0.0009
0621	43.401	0.025	10.388	0.005	1.7700	0.0009
0623	43.324	0.025	10.370	0.005	1.7668	0.0009
0624	43.446	0.025	10.399	0.005	1.7718	0.0009

[1] The certified values are traceable to the values on the respective metal certificates (EC NRM 101, NBL CRM-116 and CETAMA MP2).
[2] The certified 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.

Annex 2.GUM file for ^{242}Pu amount content determination with IDMS

Certification IRMM-049e with IRMM-1027o final

Note! that the IA input data in this file are coming from IA GUM file without uncertainty for standard IRMM-290b/A3 (K_{CRM}) and external correction (K_{ext}) (since these corections are both introduced here)! However the ratios in the blends Rb are corrected here for the uncertainty of the standard IRMM-290b/A3 (K_{CRM}) and the (K_{ext}).

ATTENTION! a correction factor K_y has been introduced in formula for average of 10 C242Pu values in order to take into account the uncertainty of the spike IRMM-1027o

ATTENTION! Introduced a new parameter C242PuCRM to take into account the uncertainty from homogeneity on 242Pu amount (parameter C_{HOM242})

Note! In this file a consistency check was applied and the delta values were modified (taking into account uncertainty for delta values of 140E-12, k = 2) according to "Consistency check" by Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x

Elements: Pu

Simplified IDMS equation for 242/239 ratio, sample (Pu-242), spike (Pu-239)

DATA FOLDER:

Sample IRMM-049e to be certified content

Spike IRMM-1027o data from certificate, reference/input values on 1/11/2012

IRMM-049e used: 7-17-24-28-37 (first series) (IDMS 18/07/2016), 53-55-66-72-81 (second series)(IDMS 08/08/2016) ATTENTION! Certification IA IRMM-049e, date of IA analysis (IA data were measured on 11/08/2016 and 24/08/2016 for 238/242, all the other ratios were remeasured on 22-23/09/2016), all ratios were decayed to a reference date 1 January 2017 and averaged

IRMM-1027o spikes used: 433-444-455-466-474 (first series), 578-599-610- 615-620 (second series)

Date of measurements: July and August 2016

Date of reference: 1 January 2017

Mass metrology certificate : E3875

Report of analysis: E3593

Atomic mass: M. Wang et al: The AME2012 atomic mass evaluation (II). Tables, graphs and references

Half-lives: Monographie BIPM-5 Table of Radionuclides, Vol.5 and 241Pu half-life: Wellum et al. 2009 (JAAS)

Model Equation:

{-------------simplified IDMS equations for 242/239 ratios----------------------}

{--------------------------------concentration calculations for 1st series and 2nd series on IDMS analysis dates ---}

$f_{c239Pu1}(m_x,n_ydec,R_b)=n_ydec/m_x *(R_ydec1-K_{CRM}*K_{ext}*R_b) / (K_{CRM}*K_{ext}*R_b-R_xdec1)*(R_xdec1);$

$c_{x1}=f_{c239Pu1}(m_{x1},n_{y1}dec,R_{b1});$
$c_{x2}=f_{c239Pu1}(m_{x2},n_{y2}dec,R_{b2});$
$c_{x3}=f_{c239Pu1}(m_{x3},n_{y3}dec,R_{b3});$
$c_{x4}=f_{c239Pu1}(m_{x4},n_{y4}dec,R_{b4});$
$c_{x5}=f_{c239Pu1}(m_{x5},n_{y5}dec,R_{b5});$

$f_{c239Pu2}(m_x,n_ydec,R_b)=n_ydec/m_x *(R_ydec2-K_{CRM}*K_{ext}*R_b) / (K_{CRM}*K_{ext}*R_b-R_xdec2)*(R_xdec2);$

$c_{x6}=f_{c239Pu2}(m_{x6},n_{y6}dec,R_{b6});$

$c_{x7}=f_{c239Pu2}(m_{x7},n_{y7}dec,R_{b7});$

$c_{x8}=f_{c239Pu2}(m_{x8},n_{y8}dec,R_{b8});$

$c_{x9}=f_{c239Pu2}(m_{x9},n_{y9}dec,R_{b9});$

$c_{x10}=f_{c239Pu2}(m_{x10},n_{y10}dec,R_{b10});$

{--------------------------------R_x decayed for 1st series and 2nd series to the IDMS analysis dates --}

$R_xdec1= Rd_{242/239} \cdot(e\text{^}(\lambda_{242}\cdot\Delta t_1)/e\text{^}(\lambda_{239}\cdot\Delta t_1))*K_{CRM}*K_{ext};$

$R_xdec2= Rd_{242/239} \cdot(e\text{^}(\lambda_{242}\cdot\Delta t_2)/e\text{^}(\lambda_{239}\cdot\Delta t_2))*K_{CRM}*K_{ext};$

{------------------------------spike parameters (here IRMM-1027o) on IDMS analysis dates---}

$R_ydec1 = R_y *(e\text{^}(-\lambda_{242}\cdot\Delta t_{spike1}))/(e\text{^}(-\lambda_{239}\cdot\Delta t_{spike1}));$

$n_{y1}dec = n_{y1} * e\text{^}(-\lambda_{239}\cdot\Delta t_{spike1})/M_{239Pu};$

$n_{y2}dec = n_{y2} * e\text{^}(-\lambda_{239}\cdot\Delta t_{spike1})/M_{239Pu};$

$n_{y3}dec = n_{y3} * e\text{^}(-\lambda_{239}\cdot\Delta t_{spike1})/M_{239Pu};$

$n_{y4}dec = n_{y4} * e\text{^}(-\lambda_{239}\cdot\Delta t_{spike1})/M_{239Pu};$

$n_{y5}dec = n_{y5} * e\text{^}(-\lambda_{239}\cdot\Delta t_{spike1})/M_{239Pu};$

$R_ydec2 = R_y *(e\text{^}(-\lambda_{242}\cdot\Delta t_{spike2}))/(e\text{^}(-\lambda_{239}\cdot\Delta t_{spike2}));$

$n_{y6}dec = n_{y6} * e\text{^}(-\lambda_{239}\cdot\Delta t_{spike2})/M_{239Pu};$

$n_{y7}dec = n_{y7} * e\text{^}(-\lambda_{239}\cdot\Delta t_{spike2})/M_{239Pu};$

$n_{y8}dec = n_{y8} * e\text{^}(-\lambda_{239}\cdot\Delta t_{spike2})/M_{239Pu};$

$n_{y9}dec = n_{y9} * e\text{^}(-\lambda_{239}\cdot\Delta t_{spike2})/M_{239Pu};$

$n_{y10}dec = n_{y10} * e\text{^}(-\lambda_{239}\cdot\Delta t_{spike2})/M_{239Pu};$

{--------------------------------concentration calculations to reference date 01/01/2017--}

$f_{c242Pu}(c_x)=c_x;$

$c_{242Pu1}=f_{c242Pu}(c_{x1})*\ e\text{^}(-\lambda_{242}\cdot\Delta t_1)+\delta_1;$

$c_{242Pu2}=f_{c242Pu}(c_{x2})*\ e\text{^}(-\lambda_{242}\cdot\Delta t_1)+\delta_2;$

$c_{242Pu3}=f_{c242Pu}(c_{x3})*\ e\text{^}(-\lambda_{242}\cdot\Delta t_1)+\delta_3;$

$c_{242Pu4}=f_{c242Pu}(c_{x4})*\ e\text{^}(-\lambda_{242}\cdot\Delta t_1)+\delta_4;$

$c_{242Pu5}=f_{c242Pu}(c_{x5})*\ e\text{^}(-\lambda_{242}\cdot\Delta t_1)+\delta_5;$

$c_{242Pu6}=f_{c242Pu}(c_{x6})*\ e\text{^}(-\lambda_{242}\cdot\Delta t_2)+\delta_6;$

$c_{242Pu7}=f_{c242Pu}(c_{x7})*\ e\text{^}(-\lambda_{242}\cdot\Delta t_2)+\delta_7;$

$c_{242Pu8}=f_{c242Pu}(c_{x8})*\ e\text{^}(-\lambda_{242}\cdot\Delta t_2)+\delta_8;$

$c_{242Pu9}=f_{c242Pu}(c_{x9})*\ e\text{^}(-\lambda_{242}\cdot\Delta t_2)+\delta_9;$

$c_{242Pu10}=f_{c242Pu}(c_{x10})*\ e\text{^}(-\lambda_{242}\cdot\Delta t_2)+\delta_{10};$

{--------------------------------concentration calculations---}

$c_{242Pu}=((c_{242Pu1}+c_{242Pu2}+c_{242Pu3}+c_{242Pu4}+c_{242Pu5}+c_{242Pu6}+c_{242Pu7}+c_{242Pu8}+c_{242Pu9}+c_{242Pu10})/10);$

$c_{242PuCRM}=((c_{242Pu1}+c_{242Pu2}+c_{242Pu3}+c_{242Pu4}+c_{242Pu5}+c_{242Pu6}+c_{242Pu7}+c_{242Pu8}+c_{242Pu9}+c_{242Pu10})/10)*K_y+c_{HOM242Pu}$

$c_{Pu}=c_{242PuCRM}/fd_{242};$

$\gamma_{242Pu}=c_{242PuCRM}*M_{242Pu};$

$\gamma_{Pu}=Md_{Pu}*c_{Pu};$

{--------------------------------operator---}

$\varepsilon_1 = c_{242Pu} - c_{242Pu1};$

$\varepsilon_2 = c_{242Pu} - c_{242Pu2};$

$\varepsilon_3 = c_{242Pu} - c_{242Pu3};$

$\varepsilon_4 = c_{242Pu} - c_{242Pu4};$

$\varepsilon_5 = c_{242Pu} - c_{242Pu5};$

$\varepsilon_6 = c_{242Pu} - c_{242Pu6};$

$\varepsilon_7 = c_{242Pu} - c_{242Pu7};$

$\varepsilon_8 = c_{242Pu} - c_{242Pu8};$

$\varepsilon_9 = c_{242Pu} - c_{242Pu9};$

$\varepsilon_{10} = c_{242Pu} - c_{242Pu10};$

{------------------- parameters for decay constants------------------}

$ln_2 = ln(2);$

$\lambda_{238} = ln_2 / \tau_{238};$

$\lambda_{239} = ln_2 / \tau_{239};$

$\lambda_{240} = ln_2 / \tau_{240};$

$\lambda_{241} = ln_2 / \tau_{241};$

$\lambda_{242} = ln_2 / \tau_{242};$

$\lambda_{244} = ln_2 / \tau_{244};$

List of Quantities:

Quantity	Unit	Definition
c_{242Pu}	mol/g	amount content of ^{242}Pu in sample (here IRMM-049e)
c_{242Pu1}	mol/g	amount content 242Pu in blend 1
$c_{242Pu10}$	mol/g	amount content 242Pu in blend 10
c_{242Pu2}	mol/g	amount content 242Pu in blend 2
c_{242Pu3}	mol/g	amount content 242Pu in blend 3
c_{242Pu4}	mol/g	amount content 242Pu in blend 4
c_{242Pu5}	mol/g	amount content 242Pu in blend 5
c_{242Pu6}	mol/g	amount content 242Pu in blend 6
c_{242Pu7}	mol/g	amount content 242Pu in blend 7
c_{242Pu8}	mol/g	amount content 242Pu in blend 8
c_{242Pu9}	mol/g	amount content 242Pu in blend 9
$c_{242PuCRM}$	mol/g	amount content of ^{242}Pu with full uncertainty
$c_{HOM242Pu}$	mol/g	amount content of ^{242}Pu homogeneity contribution

Quantity	Unit	Definition
c_{Pu}	mol/g	amount content of total Pu in sample (here IRMM-049e)
c_{x1}	mol/g	amount content of ^{242}Pu in blend 049e -1027o-07
c_{x10}	mol/g	amount content of ^{242}Pu in blend 049e -1027o-81
c_{x2}	mol/g	amount content of ^{242}Pu in blend 049e -1027o-17
c_{x3}	mol/g	amount content of ^{242}Pu in blend 049e -1027o-24
c_{x4}	mol/g	amount content of ^{242}Pu in blend 049e -1027o-28
c_{x5}	mol/g	amount content of ^{242}Pu in blend 049e -1027o-37
c_{x6}	mol/g	amount content of ^{242}Pu in blend 049e -1027o-53
c_{x7}	mol/g	amount content of ^{242}Pu in blend 049e -1027o-55
c_{x8}	mol/g	amount content of ^{242}Pu in blend 049e -1027o-66
c_{x9}	mol/g	amount content of ^{242}Pu in blend 049e -1027o-72
e		exponential
fd_{242}	mol/mol	decayed mole fraction of ^{242}Pu (on reference date)
K_{CRM}		dummy factor of 1 but with uncertainty corresponding to IRMM290/A3
K_{ext}		dummy factor of 1 but with uncertainty corresponding to external correction Excel
K_y		dummy factor of 1 but with uncertainty corresponding to the spike IRMM-1027o
ln_2		constant
M_{239Pu}	g/mol	atomic mass for ^{239}Pu
M_{242Pu}	g/mol	atomic mass for ^{242}Pu
Md_{Pu}	g/mol	molar mass of Pu on reference date 01/01/2017
m_{x1}	g	mass of sample 049e in prepared blend 1
m_{x10}	g	mass of sample 049e in prepared blend 10
m_{x2}	g	mass of sample 049e in prepared blend 2
m_{x3}	g	mass of sample 049e in prepared blend 3
m_{x4}	g	mass of sample 049e in prepared blend 4
m_{x5}	g	mass of sample 049e in prepared blend 5
m_{x6}	g	mass of sample 049e in prepared blend 6
m_{x7}	g	mass of sample 049e in prepared blend 7
m_{x8}	g	mass of sample 049e in prepared blend 8
m_{x9}	g	mass of sample 049e in prepared blend 9
n_{y1}	g	amount content of 239Pu in spike IRMM-1027o aliquot1
n_{y10}	g	amount content of 239Pu in spike IRMM-1027o aliquot10
$n_{y10}dec$	mol	decayed amount in mol of 239Pu in spike IRMM-1027o aliquot10
$n_{y1}dec$	mol	decayed amount in mol of 239Pu in spike IRMM-1027o aliquot1
n_{y2}	g	amount content of 239Pu in spike IRMM-1027o aliquot2
$n_{y2}dec$	mol	decayed amount in mol of 239Pu in spike IRMM-1027o aliquot2
n_{y3}	g	amount content of 239Pu in spike IRMM-1027o aliquot3
$n_{y3}dec$	mol	decayed amount in mol of 239Pu in spike IRMM-1027o aliquot3

Quantity	Unit	Definition
n_{y4}	g	amount content of 239Pu in spike IRMM-1027o aliquot4
n_{y4}dec	mol	decayed amount in mol of 239Pu in spike IRMM-1027o aliquot4
n_{y5}	g	amount content of 239Pu in spike IRMM-1027o aliquot5
n_{y5}dec	mol	decayed amount in mol of 239Pu in spike IRMM-1027o aliquot5
n_{y6}	g	amount content of 239Pu in spike IRMM-1027o aliquot6
n_{y6}dec	mol	decayed amount in mol of 239Pu in spike IRMM-1027o aliquot6
n_{y7}	g	amount content of 239Pu in spike IRMM-1027o aliquot7
n_{y7}dec	mol	decayed amount in mol of 239Pu in spike IRMM-1027o aliquot7
n_{y8}	g	amount content of 239Pu in spike IRMM-1027o aliquot8
n_{y8}dec	mol	decayed amount in mol of 239Pu in spike IRMM-1027o aliquot8
n_{y9}	g	amount content of 239Pu in spike IRMM-1027o aliquot9
n_{y9}dec	mol	decayed amount in mol of 239Pu in spike IRMM-1027o aliquot9
R_{b1}	mol/mol	measured $^{242}Pu/^{239}Pu$ ratio in blend 1
R_{b10}	mol/mol	measured $^{242}Pu/^{239}Pu$ ratio in blend 10
R_{b2}	mol/mol	measured $^{242}Pu/^{239}Pu$ ratio in blend 2
R_{b3}	mol/mol	measured $^{242}Pu/^{239}Pu$ ratio in blend 3
R_{b4}	mol/mol	measured $^{242}Pu/^{239}Pu$ ratio in blend 4
R_{b5}	mol/mol	measured $^{242}Pu/^{239}Pu$ ratio in blend 5
R_{b6}	mol/mol	measured $^{242}Pu/^{239}Pu$ ratio in blend 6
R_{b7}	mol/mol	measured $^{242}Pu/^{239}Pu$ ratio in blend 7
R_{b8}	mol/mol	measured $^{242}Pu/^{239}Pu$ ratio in blend 8
R_{b9}	mol/mol	measured $^{242}Pu/^{239}Pu$ ratio in blend 9
$Rd_{242/239}$	mol/mol	isotope amount ratio n_{242}/n_{239} after decay to reference date
$R_x$dec1	mol/mol	decayed isotope amount ratio n_{242}/n_{239} as from the IA measurements for IRMM-049e (IA GUM FILE) 1st series
$R_x$dec2	mol/mol	decayed isotope amount ratio n_{242}/n_{239} as from the IA measurements for IRMM-049e (IA GUM FILE) 2nd series
R_y	mol/mol	isotope amount ratio n_{242}/n_{239} as given in the spike certificate (IRMM-1027o)
$R_y$dec1	mol/mol	decayed isotope amount ratio n_{242}/n_{239} as given in the spike certificate (IRMM-1027o) 1st series
$R_y$dec2	mol/mol	decayed isotope amount ratio n_{242}/n_{239} as given in the spike certificate (IRMM-1027o) 2nd series
δ_1		parameter of consistency check
δ_{10}		parameter of consistency check
δ_2		parameter of consistency check
δ_3		parameter of consistency check
δ_4		parameter of consistency check
δ_5		parameter of consistency check
δ_6		parameter of consistency check

Quantity	Unit	Definition
δ_7		parameter of consistency check
δ_8		parameter of consistency check
δ_9		parameter of consistency check
Δt_1	a	time difference measurement date 18 July 2016 1st IDMS series and reference date 1 January 2017
Δt_2	a	time difference measurement date 08 August 2016 2nd IDMS series and reference date 1 January 2017
Δt_{spike1}	a	time difference between reference date of spike 01/11/2012 and measurement date 18 July 2016 1st IDMS series
Δt_{spike2}	a	time difference between reference date of spike 01/11/2012 and measurement date 08 August 2016 2nd IDMS series
ε_1		see in Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x
ε_{10}		see in Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x
ε_2		see in Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x
ε_3		see in Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x
ε_4		see in Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x
ε_5		see in Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x
ε_6		see in Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x
ε_7		see in Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x
ε_8		see in Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x
ε_9		see in Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x
γ_{242Pu}	g/g	mass content of ^{242}Pu in sample (here IRMM-049e)
γ_{Pu}	g/g	mass content of total Pu in sample (here IRMM-049e)
λ_{238}	a^{-1}	decay constant ^{238}Pu
λ_{239}	a^{-1}	decay constant ^{239}Pu
λ_{240}	a^{-1}	decay constant ^{240}Pu
λ_{241}	a^{-1}	decay constant ^{241}Pu
λ_{242}	a^{-1}	decay constant ^{242}Pu
λ_{244}	a^{-1}	decay constant ^{244}Pu
τ_{238}	a	half life ^{238}Pu
τ_{239}	a	half life ^{239}Pu
τ_{240}	a	half life ^{240}Pu
τ_{241}	a	half life ^{241}Pu
τ_{242}	a	half life ^{242}Pu
τ_{244}	a	half life ^{244}Pu

$c_{HOM242Pu}$: Type B normal distribution
Value: 0 mol/g
Expanded Uncertainty: $6.6326 \cdot 10^{-11}$ mol/g
Coverage Factor: 1

Expanded Uncertainty is the ABSOLUTE uncertainty of the homogeneity contribution, calcualted as c_{242Pu}*ubb,rel, 6.6326E-11

e: Constant
Value: 2.71828182845904523536

fd_{242}: Import
Filename: Certification IA IRMM-049e final-v5-4-with uncertainty std.SMU
Symbol: f_{242}

amount abundance for 242 decayed to reference date 01/01/2017 with full uncertainty (including U-CRM)

K_{CRM}: Type B normal distribution
Value: 1
Expanded Uncertainty: 0.1 %
Coverage Factor: 2

uncertainty of standard IRMM-290b/A3 (K-factor) introduced here instead of in the measurement file (Excel) to take it into account, for 3 mass units

K_{ext}: Type B normal distribution
Value: 1
Expanded Uncertainty: 0.01 %
Coverage Factor: 1

uncertainty of external correction (K-factor) introduced here instead of in the measurement file (Excel) to take it into account, 0.01%, k=1

K_y: Type B normal distribution
Value: 1
Expanded Uncertainty: 0.0006
Coverage Factor: 2

uncertainty of the spike (IRMM-1027o) introduced here to take it into account, should be the (typical) relative uncertainty of $n_{y1,}$ $n_{y2...}$ 0.06%

M_{239Pu}: Type B normal distribution
Value: 239.0521636 g/mol
Expanded Uncertainty: 0.0000038 g/mol
Coverage Factor: 2

M. Wang et al: The AME2012 atomic mass evaluation (II)

M_{242Pu}: Type B normal distribution
Value: 242.0587428 g/mol
Expanded Uncertainty: 0.0000040 g/mol
Coverage Factor: 2

M. Wang et al: The AME2012 atomic mass evaluation (II)

Md_{Pu}: Import
Filename: Certification IA IRMM-049e final-v5-4-with uncertainty std.SMU
Symbol: M_{Pu}

total mass fraction of Pu decayed to reference date 01/01/2017 with full uncertainty (including U_{CRM})

m_{x1}: Type B normal distribution
Value: 5.99902 g
Expanded Uncertainty: 0.00009 g
Coverage Factor: 2

Weighing certificate E3875 unit#7

m_{x10}: Type B normal distribution
Value: 6.03342 g
Expanded Uncertainty: 0.00010 g
Coverage Factor: 2

Weighing certificate E3875 unit#81

m_{x2}: Type B normal distribution
Value: 5.93325 g
Expanded Uncertainty: 0.00008 g
Coverage Factor: 2

Weighing certificate E3875 unit#17

m_{x3}: Type B normal distribution
Value: 6.01834 g
Expanded Uncertainty: 0.00008 g
Coverage Factor: 2

Weighing certificate E3875 unit#24

m_{x4}: Type B normal distribution
Value: 5.97342 g
Expanded Uncertainty: 0.00012 g
Coverage Factor: 2

Weighing certificate E3875 unit#28

m_{x5}: Type B normal distribution
Value: 6.18060 g
Expanded Uncertainty: 0.00008 g
Coverage Factor: 2

Weighing certificate E3875 unit#37

m_{x6}: Type B normal distribution
Value: 6.00926 g
Expanded Uncertainty: 0.00012 g
Coverage Factor: 2

Weighing certificate E3875 unit#53

m_{x7}: Type B normal distribution
Value: 5.99841 g
Expanded Uncertainty: 0.00008 g
Coverage Factor: 2

Weighing certificate E3875 unit#55

m_{x8}: Type B normal distribution
Value: 6.02214 g
Expanded Uncertainty: 0.00011 g
Coverage Factor: 2

Weighing certificate E3875 unit#66

m_{x9}: Type B normal distribution
Value: 5.93003 g
Expanded Uncertainty: 0.00010 g
Coverage Factor: 2

Weighing certificate E3875 unit#72

n_{y1}: Type B normal distribution
Value: $1.7694 \cdot 10^{-3}$ g
Expanded Uncertainty: 0 g
Coverage Factor: 2

IRMM-1027o N433 corrected mass = 2.5329 g

n_{y10}: Type B normal distribution
Value: $1.7667 \cdot 10^{-3}$ g
Expanded Uncertainty: 0 g
Coverage Factor: 2

IRMM-1027o N620 corrected mass =2.5290 g

n_{y2}: Type B normal distribution
Value: $1.7739 \cdot 10^{-3}$ g
Expanded Uncertainty: 0 g
Coverage Factor: 2

IRMM-1027o N444 corrected mass = 2.5394 g

n_{y3}: Type B normal distribution
Value: $1.7701 \cdot 10^{-3}$ g
Expanded Uncertainty: 0 g
Coverage Factor: 2

IRMM-1027o N455 corrected mass = 2.5339 g

n_{y4}: Type B normal distribution
Value: $1.7644 \cdot 10^{-3}$ g
Expanded Uncertainty: 0 g
Coverage Factor: 2

IRMM-1027o N466 corrected mass = 2.5257 g

n_{y5}: Type B normal distribution
Value: $1.7687 \cdot 10^{-3}$ g
Expanded Uncertainty: 0 g
Coverage Factor: 2

IRMM-1027o N474 corrected mass = 2.5319 g

n_{y6}: Type B normal distribution
Value: $1.7643 \cdot 10^{-3}$ g
Expanded Uncertainty: 0 g
Coverage Factor: 2

IRMM-1027o N578 corrected mass = 2.5256 g

n_{y7}: Type B normal distribution
Value: $1.7684 \cdot 10^{-3}$ g
Expanded Uncertainty: 0 g
Coverage Factor: 2

IRMM-1027o N599 corrected mass = 2.5315 g

n_{y8}: Type B normal distribution
Value: $1.7705 \cdot 10^{-3}$ g
Expanded Uncertainty: 0 g
Coverage Factor: 2

IRMM-1027o N610 corrected mass =2.5345 g

n_{y9}: Type B normal distribution
Value: $1.7688 \cdot 10^{-3}$ g
Expanded Uncertainty: 0 g
Coverage Factor: 2

IRMM-1027o N615 corrected mass =2.5321 g

R_{b1}: Type B normal distribution
Value: 0.290342 mol/mol
Expanded Uncertainty: $3.4 \cdot 10^{-5}$ mol/mol
Coverage Factor: 2

unit#7 P160718 IRMM-049e with IRMM1027o TurretA-2.xls

R_{b10}: Type B normal distribution
Value: 0.292454 mol/mol
Expanded Uncertainty: 0.000093 mol/mol
Coverage Factor: 2

unit#81 P160808 IRMM-049e with IRMM1027o TurretB-2.xls

R_{b2}: Type B normal distribution
Value: 0.286430 mol/mol
Expanded Uncertainty: 0.000083 mol/mol
Coverage Factor: 2

unit#17 P160718 IRMM-049e with IRMM1027o TurretA-2.xls

R_{b3}: Type B normal distribution
Value: 0.291188 mol/mol
Expanded Uncertainty: 0.000039 mol/mol
Coverage Factor: 2

unit#24 P160718 IRMM-049e with IRMM1027o TurretA-2.xls

R_{b4}: Type B normal distribution
Value: 0.28988 mol/mol
Expanded Uncertainty: 0.00014 mol/mol
Coverage Factor: 2

unit#28 P160718 IRMM-049e with IRMM1027o TurretA-2.xls

R_{b5}: Type B normal distribution
Value: 0.299083 mol/mol
Expanded Uncertainty: 0.000036 mol/mol
Coverage Factor: 2

unit#37 P160718 IRMM-049e with IRMM1027o TurretA-2.xls

R_{b6}: Type B normal distribution
Value: 0.291692 mol/mol
Expanded Uncertainty: 0.000035 mol/mol
Coverage Factor: 2

unit#53 P160808 IRMM-049e with IRMM1027o TurretB-2.xls

R_{b7}: Type B normal distribution
Value: 0.290460 mol/mol
Expanded Uncertainty: 0.000072 mol/mol
Coverage Factor: 2

unit#55 P160808 IRMM-049e with IRMM1027o TurretB-2.xls

R_{b8}: Type B normal distribution
Value: 0.291148 mol/mol
Expanded Uncertainty: 0.000036 mol/mol
Coverage Factor: 2

unit#66 P160808 IRMM-049e with IRMM1027o TurretB-2.xls

R_{b9}: Type B normal distribution
Value: 0.287016 mol/mol
Expanded Uncertainty: 0.000029 mol/mol
Coverage Factor: 2

unit#72 P160808 IRMM-049e with IRMM1027o TurretB-2.xls

$Rd_{242/239}$: Import
Filename: Certification IA IRMM-049e final-v5-without uncertainty std.SMU
Symbol: $Rd_{242/239}$

input data: Certification IA IRMM-049e final-v5-without uncertainty std.SMU ATTENTION! for these ratios the values used are those from re-measurements on turrets E and F

R_y: Type B normal distribution
Value: 0.0000757 mol/mol
Expanded Uncertainty: 0.0000008 mol/mol
Coverage Factor: 2

Ratio 242/239Pu as given in the spike certificate (IRMM-1027o)

δ_1: Type B normal distribution
Value: 0
Expanded Uncertainty: $140 \cdot 10^{-12}$
Coverage Factor: 2

Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x

δ_{10}: Type B normal distribution
Value: 0
Expanded Uncertainty: $140 \cdot 10^{-12}$
Coverage Factor: 2

Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x

δ_2: Type B normal distribution
Value: 0
Expanded Uncertainty: $140 \cdot 10^{-12}$
Coverage Factor: 2

Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x

δ_3: Type B normal distribution
Value: 0
Expanded Uncertainty: $140 \cdot 10^{-12}$
Coverage Factor: 2

Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x

δ_4: Type B normal distribution
Value: 0
Expanded Uncertainty: $140 \cdot 10^{-12}$
Coverage Factor: 2

Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x

δ_5: Type B normal distribution
Value: 0
Expanded Uncertainty: $140 \cdot 10^{-12}$
Coverage Factor: 2

Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x

δ_6: Type B normal distribution
Value: 0
Expanded Uncertainty: $140 \cdot 10^{-12}$
Coverage Factor: 2

Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x

δ_7: Type B normal distribution
Value: 0
Expanded Uncertainty: $140 \cdot 10^{-12}$
Coverage Factor: 2

Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x

δ_8: Type B normal distribution
Value: 0
Expanded Uncertainty: $140 \cdot 10^{-12}$
Coverage Factor: 2

Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x

δ_9: Type B normal distribution
Value: 0
Expanded Uncertainty: $140 \cdot 10^{-12}$
Coverage Factor: 2

Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x

Δt_1: Constant
Value: 0.457221081 a

time difference between IDMS analysis date and reference date is 0.457221081 a

Δt_2: Constant
Value: 0.399726215 a

time difference between IDMS analysis date and reference date is of 0.399726215 a

Δt_{spike1}: Constant
Value: 3.7098 a

time difference between reference date of spike 01/11/2012 and measurement date 18 July 2016 1st IDMS series = 1355d

Δt_{spike2}: Constant
Value: 3.7673 a

time difference between reference date of spike 01/11/2012 and measurement date 08 August 2016 1st IDMS series = 1376 d

τ_{236}: Type B normal distribution
Value: 87.74 a
Expanded Uncertainty: 0.03 a
Coverage Factor: 1

Monographie BIPM-5 Table of Radionuclides, Vol.5

τ_{239}: Type B normal distribution
Value: 24100 a
Expanded Uncertainty: 11 a
Coverage Factor: 1

Monographie BIPM-5 Table of Radionuclides, Vol.5

τ_{240}: Type B normal distribution
Value: 6561 a
Expanded Uncertainty: 7 a
Coverage Factor: 1

Monographie BIPM-5 Table of Radionuclides, Vol.5

τ_{241}: Type B normal distribution
Value: 14.325 a
Expanded Uncertainty: 0.024 a
Coverage Factor: 2

Wellum et al. 2009 (JAAS)

τ_{242}: Type B normal distribution
Value: 373000 a
Expanded Uncertainty: 3000 a
Coverage Factor: 1

Monographie BIPM-5 Table of Radionuclides, Vol.5

τ_{244}: Type B normal distribution
Value: $8 \cdot 10^7$ a
Expanded Uncertainty: $0.09 \cdot 10^7$ a
Coverage Factor: 1

Monographie BIPM-5 Table of Radionuclides, Vol.5

Input Correlation:

	fd_{242}	Md_{Pu}
fd_{242}	1	0.9956
Md_{Pu}	0.9956	1

The abundance set for Pu is assumed as uncorrelated.

Interim Results:

Quantity	Value	Standard Uncertainty
$n_{y10}dec$	$7.389636461 \cdot 10^{-6}$ mol	$370 \cdot 10^{-15}$ mol
$n_{y1}dec$	$7.400942082 \cdot 10^{-6}$ mol	$365 \cdot 10^{-15}$ mol
$n_{y2}dec$	$7.419764417 \cdot 10^{-6}$ mol	$366 \cdot 10^{-15}$ mol
$n_{y3}dec$	$7.403870001 \cdot 10^{-6}$ mol	$365 \cdot 10^{-15}$ mol
$n_{y4}dec$	$7.380028377 \cdot 10^{-6}$ mol	$364 \cdot 10^{-15}$ mol
$n_{y5}dec$	$7.398014164 \cdot 10^{-6}$ mol	$365 \cdot 10^{-15}$ mol
$n_{y6}dec$	$7.379597899 \cdot 10^{-6}$ mol	$370 \cdot 10^{-15}$ mol
$n_{y7}dec$	$7.396747109 \cdot 10^{-6}$ mol	$371 \cdot 10^{-15}$ mol
$n_{y8}dec$	$7.405530851 \cdot 10^{-6}$ mol	$371 \cdot 10^{-15}$ mol
$n_{y9}dec$	$7.398420202 \cdot 10^{-6}$ mol	$371 \cdot 10^{-15}$ mol
λ_{242}	$1.8583 \cdot 10^{-6}$ a^{-1}	$14.9 \cdot 10^{-9}$ a^{-1}

Uncertainty Budgets:

$c_{242PuCRM}$: amount content of ^{242}Pu with full uncertainty

Quantity	Value	Standard Uncertainty	Distribution	Sensitivity Coefficient	Uncertainty Contribution	Index
$c_{HOM242Pu}$	0.0 mol/g	$66.3\cdot10^{-12}$ mol/g	normal	1.0	$66\cdot10^{-12}$ mol/g	8.8 %
e	2.718281828459					
K_{CRM}	1.000000	$500\cdot10^{-6}$	normal	$360\cdot10^{-9}$	$180\cdot10^{-12}$ mol/g	64.2 %
K_{ext}	1.000000	$100\cdot10^{-6}$	normal	$360\cdot10^{-9}$	$36\cdot10^{-12}$ mol/g	2.6 %
K_y	1.000000	$300\cdot10^{-6}$	normal	$360\cdot10^{-9}$	$110\cdot10^{-12}$ mol/g	23.1 %
M_{239Pu}	239.05216360 g/mol	$1.90\cdot10^{-6}$ g/mol	normal	$-1.5\cdot10^{-9}$	$-2.8\cdot10^{-15}$ mol/g	0.0 %
m_{x1}	5.9990200 g	$45.0\cdot10^{-6}$ g	normal	$-6.0\cdot10^{-9}$	$-270\cdot10^{-15}$ mol/g	0.0 %
m_{x10}	6.0334200 g	$50.0\cdot10^{-6}$ g	normal	$-5.9\cdot10^{-9}$	$-300\cdot10^{-15}$ mol/g	0.0 %
m_{x2}	5.9332500 g	$40.0\cdot10^{-6}$ g	normal	$-6.0\cdot10^{-9}$	$-240\cdot10^{-15}$ mol/g	0.0 %
m_{x3}	6.0183400 g	$40.0\cdot10^{-6}$ g	normal	$-6.0\cdot10^{-9}$	$-240\cdot10^{-15}$ mol/g	0.0 %
m_{x4}	5.9734200 g	$60.0\cdot10^{-6}$ g	normal	$-6.0\cdot10^{-9}$	$-360\cdot10^{-15}$ mol/g	0.0 %
m_{x5}	6.1806000 g	$40.0\cdot10^{-6}$ g	normal	$-5.8\cdot10^{-9}$	$-230\cdot10^{-15}$ mol/g	0.0 %
m_{x6}	6.0092600 g	$60.0\cdot10^{-6}$ g	normal	$-6.0\cdot10^{-9}$	$-360\cdot10^{-15}$ mol/g	0.0 %
m_{x7}	5.9984100 g	$40.0\cdot10^{-6}$ g	normal	$-6.0\cdot10^{-9}$	$-240\cdot10^{-15}$ mol/g	0.0 %
m_{x8}	6.0221400 g	$55.0\cdot10^{-6}$ g	normal	$-5.9\cdot10^{-9}$	$-330\cdot10^{-15}$ mol/g	0.0 %
m_{x9}	5.9300300 g	$50.0\cdot10^{-6}$ g	normal	$-6.0\cdot10^{-9}$	$-300\cdot10^{-15}$ mol/g	0.0 %
n_{y1}	$1.7694\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 mol/g	0.0 %
n_{y10}	$1.7667\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 mol/g	0.0 %
n_{y2}	$1.7739\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 mol/g	0.0 %
n_{y3}	$1.7701\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 mol/g	0.0 %
n_{y4}	$1.7644\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 mol/g	0.0 %
n_{y5}	$1.7687\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 mol/g	0.0 %
n_{y6}	$1.7643\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 mol/g	0.0 %
n_{y7}	$1.7684\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 mol/g	0.0 %
n_{y8}	$1.7705\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 mol/g	0.0 %
n_{y9}	$1.7688\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 mol/g	0.0 %

Quantity	Value	Standard Uncertainty	Distribution	Sensitivity Coefficient	Uncertainty Contribution	Index
R_{b1}	0.2903420 mol/mol	$17.0\cdot10^{-6}$ mol/mol	normal	$120\cdot10^{-9}$	$2.1\cdot10^{-12}$ mol/g	0.0 %
R_{b10}	0.2924540 mol/mol	$46.5\cdot10^{-6}$ mol/mol	normal	$120\cdot10^{-9}$	$5.7\cdot10^{-12}$ mol/g	0.0 %
R_{b2}	0.2864300 mol/mol	$41.5\cdot10^{-6}$ mol/mol	normal	$130\cdot10^{-9}$	$5.2\cdot10^{-12}$ mol/g	0.0 %
R_{b3}	0.2911880 mol/mol	$19.5\cdot10^{-6}$ mol/mol	normal	$120\cdot10^{-9}$	$2.4\cdot10^{-12}$ mol/g	0.0 %
R_{b4}	0.2898800 mol/mol	$70.0\cdot10^{-6}$ mol/mol	normal	$120\cdot10^{-9}$	$8.7\cdot10^{-12}$ mol/g	0.2 %
R_{b5}	0.2990830 mol/mol	$18.0\cdot10^{-6}$ mol/mol	normal	$120\cdot10^{-9}$	$2.2\cdot10^{-12}$ mol/g	0.0 %
R_{b6}	0.2916920 mol/mol	$17.5\cdot10^{-6}$ mol/mol	normal	$120\cdot10^{-9}$	$2.2\cdot10^{-12}$ mol/g	0.0 %
R_{b7}	0.2904600 mol/mol	$36.0\cdot10^{-6}$ mol/mol	normal	$120\cdot10^{-9}$	$4.4\cdot10^{-12}$ mol/g	0.0 %
R_{b8}	0.2911480 mol/mol	$18.0\cdot10^{-6}$ mol/mol	normal	$120\cdot10^{-9}$	$2.2\cdot10^{-12}$ mol/g	0.0 %
R_{b9}	0.2870160 mol/mol	$14.5\cdot10^{-6}$ mol/mol	normal	$120\cdot10^{-9}$	$1.8\cdot10^{-12}$ mol/g	0.0 %
$Rd_{242/239}$	450.085 mol/mol	0.121 mol/mol		$-510\cdot10^{-15}$	$-62\cdot10^{-15}$ mol/g	0.0 %
R_y	$75.700\cdot10^{-6}$ mol/mol	$400\cdot10^{-9}$ mol/mol	normal	$-1.2\cdot10^{-6}$	$-490\cdot10^{-15}$ mol/g	0.0 %
δ_1	0.0	$70.0\cdot10^{-12}$	normal	0.10	$7.0\cdot10^{-12}$ mol/g	0.0 %
δ_{10}	0.0	$70.0\cdot10^{-12}$	normal	0.10	$7.0\cdot10^{-12}$ mol/g	0.0 %
δ_2	0.0	$70.0\cdot10^{-12}$	normal	0.10	$7.0\cdot10^{-12}$ mol/g	0.0 %
δ_3	0.0	$70.0\cdot10^{-12}$	normal	0.10	$7.0\cdot10^{-12}$ mol/g	0.0 %
δ_4	0.0	$70.0\cdot10^{-12}$	normal	0.10	$7.0\cdot10^{-12}$ mol/g	0.0 %
δ_5	0.0	$70.0\cdot10^{-12}$	normal	0.10	$7.0\cdot10^{-12}$ mol/g	0.0 %
δ_6	0.0	$70.0\cdot10^{-12}$	normal	0.10	$7.0\cdot10^{-12}$ mol/g	0.0 %
δ_7	0.0	$70.0\cdot10^{-12}$	normal	0.10	$7.0\cdot10^{-12}$ mol/g	0.0 %
δ_8	0.0	$70.0\cdot10^{-12}$	normal	0.10	$7.0\cdot10^{-12}$ mol/g	0.0 %
δ_9	0.0	$70.0\cdot10^{-12}$	normal	0.10	$7.0\cdot10^{-12}$ mol/g	0.0 %
Δt_1	0.457221081 a					
Δt_2	0.399726215 a					
Δt_{spike1}	3.7098 a					
Δt_{spike2}	3.7673 a					
τ_{239}	24100.0 a	11.0 a	normal	$1.6\cdot10^{-15}$	$18\cdot10^{-15}$ mol/g	0.0 %
τ_{242}	$373.00\cdot10^{3}$ a	3000 a	normal	$760\cdot10^{-21}$	$2.3\cdot10^{-15}$ mol/g	0.0 %
$c_{242PuCRM}$	$358.282\cdot10^{-9}$ mol/g	$224\cdot10^{-12}$ mol/g				

c_{Pu}: **amount content of total Pu in sample (here IRMM-049e)**

Quantity	Value	Standard Uncertainty	Distribution	Sensitivity Coefficient	Uncertainty Contribution	Index
$c_{HOM242Pu}$	0.0 mol/g	$66.3\cdot10^{-12}$ mol/g	normal	1.1	$70\cdot10^{-12}$ mol/g	8.8 %
e	2.718281828459					
fd_{242}	0.9471745 mol/mol	$25.0\cdot10^{-6}$ mol/mol		$-400\cdot10^{-9}$	$-10\cdot10^{-12}$ mol/g	0.2 %
K_{CRM}	1.000000	$500\cdot10^{-6}$	normal	$380\cdot10^{-9}$	$190\cdot10^{-12}$ mol/g	64.1 %
K_{ext}	1.000000	$100\cdot10^{-6}$	normal	$380\cdot10^{-9}$	$38\cdot10^{-12}$ mol/g	2.6 %
K_y	1.000000	$300\cdot10^{-6}$	normal	$380\cdot10^{-9}$	$110\cdot10^{-12}$ mol/g	23.1 %
M_{239Pu}	239.05216360 g/mol	$1.90\cdot10^{-6}$ g/mol	normal	$-1.6\cdot10^{-9}$	$-3.0\cdot10^{-15}$ mol/g	0.0 %
m_{x1}	5.9990200 g	$45.0\cdot10^{-6}$ g	normal	$-6.3\cdot10^{-9}$	$-280\cdot10^{-15}$ mol/g	0.0 %
m_{x10}	6.0334200 g	$50.0\cdot10^{-6}$ g	normal	$-6.3\cdot10^{-9}$	$-310\cdot10^{-15}$ mol/g	0.0 %
m_{x2}	5.9332500 g	$40.0\cdot10^{-6}$ g	normal	$-6.4\cdot10^{-9}$	$-260\cdot10^{-15}$ mol/g	0.0 %
m_{x3}	6.0183400 g	$40.0\cdot10^{-6}$ g	normal	$-6.3\cdot10^{-9}$	$-250\cdot10^{-15}$ mol/g	0.0 %
m_{x4}	5.9734200 g	$60.0\cdot10^{-6}$ g	normal	$-6.3\cdot10^{-9}$	$-380\cdot10^{-15}$ mol/g	0.0 %
m_{x5}	6.1806000 g	$40.0\cdot10^{-6}$ g	normal	$-6.1\cdot10^{-9}$	$-240\cdot10^{-15}$ mol/g	0.0 %
m_{x6}	6.0092600 g	$60.0\cdot10^{-6}$ g	normal	$-6.3\cdot10^{-9}$	$-380\cdot10^{-15}$ mol/g	0.0 %
m_{x7}	5.9984100 g	$40.0\cdot10^{-6}$ g	normal	$-6.3\cdot10^{-9}$	$-250\cdot10^{-15}$ mol/g	0.0 %
m_{x8}	6.0221400 g	$55.0\cdot10^{-6}$ g	normal	$-6.3\cdot10^{-9}$	$-350\cdot10^{-15}$ mol/g	0.0 %
m_{x9}	5.9300300 g	$50.0\cdot10^{-6}$ g	normal	$-6.4\cdot10^{-9}$	$-320\cdot10^{-15}$ mol/g	0.0 %
n_{y1}	$1.7694\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 mol/g	0.0 %
n_{y10}	$1.7667\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 mol/g	0.0 %
n_{y2}	$1.7739\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 mol/g	0.0 %
n_{y3}	$1.7701\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 mol/g	0.0 %
n_{y4}	$1.7644\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 mol/g	0.0 %
n_{y5}	$1.7687\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 mol/g	0.0 %
n_{y6}	$1.7643\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 mol/g	0.0 %
n_{y7}	$1.7684\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 mol/g	0.0 %
n_{y8}	$1.7705\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 mol/g	0.0 %
n_{y9}	$1.7688\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 mol/g	0.0 %

Quantity	Value	Standard Uncertainty	Distribution	Sensitivity Coefficient	Uncertainty Contribution	Index
R_{b1}	0.2903420 mol/mol	$17.0\cdot10^{-6}$ mol/mol	normal	$130\cdot10^{-9}$	$2.2\cdot10^{-12}$ mol/g	0.0 %
R_{b10}	0.2924540 mol/mol	$46.5\cdot10^{-6}$ mol/mol	normal	$130\cdot10^{-9}$	$6.0\cdot10^{-12}$ mol/g	0.0 %
R_{b2}	0.2864300 mol/mol	$41.5\cdot10^{-6}$ mol/mol	normal	$130\cdot10^{-9}$	$5.5\cdot10^{-12}$ mol/g	0.0 %
R_{b3}	0.2911880 mol/mol	$19.5\cdot10^{-6}$ mol/mol	normal	$130\cdot10^{-9}$	$2.5\cdot10^{-12}$ mol/g	0.0 %
R_{b4}	0.2898800 mol/mol	$70.0\cdot10^{-6}$ mol/mol	normal	$130\cdot10^{-9}$	$9.1\cdot10^{-12}$ mol/g	0.2 %
R_{b5}	0.2990830 mol/mol	$18.0\cdot10^{-6}$ mol/mol	normal	$130\cdot10^{-9}$	$2.3\cdot10^{-12}$ mol/g	0.0 %
R_{b6}	0.2916920 mol/mol	$17.5\cdot10^{-6}$ mol/mol	normal	$130\cdot10^{-9}$	$2.3\cdot10^{-12}$ mol/g	0.0 %
R_{b7}	0.2904600 mol/mol	$36.0\cdot10^{-6}$ mol/mol	normal	$130\cdot10^{-9}$	$4.7\cdot10^{-12}$ mol/g	0.0 %
R_{b8}	0.2911480 mol/mol	$18.0\cdot10^{-6}$ mol/mol	normal	$130\cdot10^{-9}$	$2.3\cdot10^{-12}$ mol/g	0.0 %
R_{b9}	0.2870160 mol/mol	$14.5\cdot10^{-6}$ mol/mol	normal	$130\cdot10^{-9}$	$1.9\cdot10^{-12}$ mol/g	0.0 %
$Rd_{242/239}$	450.085 mol/mol	0.121 mol/mol		$-540\cdot10^{-15}$	$-66\cdot10^{-15}$ mol/g	0.0 %
R_y	$75.700\cdot10^{-6}$ mol/mol	$400\cdot10^{-9}$ mol/mol	normal	$-1.3\cdot10^{-6}$	$-520\cdot10^{-15}$ mol/g	0.0 %
δ_1	0.0	$70.0\cdot10^{-12}$	normal	0.11	$7.4\cdot10^{-12}$ mol/g	0.0 %
δ_{10}	0.0	$70.0\cdot10^{-12}$	normal	0.11	$7.4\cdot10^{-12}$ mol/g	0.0 %
δ_2	0.0	$70.0\cdot10^{-12}$	normal	0.11	$7.4\cdot10^{-12}$ mol/g	0.0 %
δ_3	0.0	$70.0\cdot10^{-12}$	normal	0.11	$7.4\cdot10^{-12}$ mol/g	0.0 %
δ_4	0.0	$70.0\cdot10^{-12}$	normal	0.11	$7.4\cdot10^{-12}$ mol/g	0.0 %
δ_5	0.0	$70.0\cdot10^{-12}$	normal	0.11	$7.4\cdot10^{-12}$ mol/g	0.0 %
δ_6	0.0	$70.0\cdot10^{-12}$	normal	0.11	$7.4\cdot10^{-12}$ mol/g	0.0 %
δ_7	0.0	$70.0\cdot10^{-12}$	normal	0.11	$7.4\cdot10^{-12}$ mol/g	0.0 %
δ_8	0.0	$70.0\cdot10^{-12}$	normal	0.11	$7.4\cdot10^{-12}$ mol/g	0.0 %
δ_9	0.0	$70.0\cdot10^{-12}$	normal	0.11	$7.4\cdot10^{-12}$ mol/g	0.0 %
Δt_1	0.457221081 a					
Δt_2	0.399726215 a					
Δt_{spike1}	3.7098 a					
Δt_{spike2}	3.7673 a					
τ_{239}	24100.0 a	11.0 a	normal	$1.7\cdot10^{-15}$	$19\cdot10^{-15}$ mol/g	0.0 %
τ_{242}	$373.00\cdot10^{3}$ a	3000 a	normal	$810\cdot10^{-21}$	$2.4\cdot10^{-15}$ mol/g	0.0 %
c_{Pu}	$378.263\cdot10^{-9}$ mol/g	$236\cdot10^{-12}$ mol/g				

y_{242Pu}: mass content of ^{242}Pu in sample (here IRMM-049e)

Quantity	Value	Standard Uncertainty	Distribution	Sensitivity Coefficient	Uncertainty Contribution	Index
$c_{HOM242Pu}$	0.0 mol/g	$66.3\cdot10^{-12}$ mol/g	normal	240	$16\cdot10^{-9}$ g/g	8.8 %
e	2.718281828459					
K_{CRM}	1.000000	$500\cdot10^{-6}$	normal	$87\cdot10^{-6}$	$43\cdot10^{-9}$ g/g	64.2 %
K_{ext}	1.000000	$100\cdot10^{-6}$	normal	$87\cdot10^{-6}$	$8.7\cdot10^{-9}$ g/g	2.6 %
K_y	1.000000	$300\cdot10^{-6}$	normal	$87\cdot10^{-6}$	$26\cdot10^{-9}$ g/g	23.1 %
M_{239Pu}	239.05216360 g/mol	$1.90\cdot10^{-6}$ g/mol	normal	$-360\cdot10^{-9}$	$-690\cdot10^{-15}$ g/g	0.0 %
M_{242Pu}	242.05874280 g/mol	$2.00\cdot10^{-6}$ g/mol	normal	$360\cdot10^{-9}$	$720\cdot10^{-15}$ g/g	0.0 %
m_{x1}	5.9990200 g	$45.0\cdot10^{-6}$ g	normal	$-1.4\cdot10^{-6}$	$-65\cdot10^{-12}$ g/g	0.0 %
m_{x10}	6.0334200 g	$50.0\cdot10^{-6}$ g	normal	$-1.4\cdot10^{-6}$	$-72\cdot10^{-12}$ g/g	0.0 %
m_{x2}	5.9332500 g	$40.0\cdot10^{-6}$ g	normal	$-1.5\cdot10^{-6}$	$-58\cdot10^{-12}$ g/g	0.0 %
m_{x3}	6.0183400 g	$40.0\cdot10^{-6}$ g	normal	$-1.4\cdot10^{-6}$	$-58\cdot10^{-12}$ g/g	0.0 %
m_{x4}	5.9734200 g	$60.0\cdot10^{-6}$ g	normal	$-1.5\cdot10^{-6}$	$-87\cdot10^{-12}$ g/g	0.0 %
m_{x5}	6.1806000 g	$40.0\cdot10^{-6}$ g	normal	$-1.4\cdot10^{-6}$	$-56\cdot10^{-12}$ g/g	0.0 %
m_{x6}	6.0092600 g	$60.0\cdot10^{-6}$ g	normal	$-1.4\cdot10^{-6}$	$-87\cdot10^{-12}$ g/g	0.0 %
m_{x7}	5.9984100 g	$40.0\cdot10^{-6}$ g	normal	$-1.4\cdot10^{-6}$	$-58\cdot10^{-12}$ g/g	0.0 %
m_{x8}	6.0221400 g	$55.0\cdot10^{-6}$ g	normal	$-1.4\cdot10^{-6}$	$-79\cdot10^{-12}$ g/g	0.0 %
m_{x9}	5.9300300 g	$50.0\cdot10^{-6}$ g	normal	$-1.5\cdot10^{-6}$	$-73\cdot10^{-12}$ g/g	0.0 %
n_{y1}	$1.7694\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 g/g	0.0 %
n_{y10}	$1.7667\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 g/g	0.0 %
n_{y2}	$1.7739\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 g/g	0.0 %
n_{y3}	$1.7701\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 g/g	0.0 %
n_{y4}	$1.7644\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 g/g	0.0 %
n_{y5}	$1.7687\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 g/g	0.0 %
n_{y6}	$1.7643\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 g/g	0.0 %
n_{y7}	$1.7684\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 g/g	0.0 %
n_{y8}	$1.7705\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 g/g	0.0 %
n_{y9}	$1.7688\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 g/g	0.0 %
R_{b1}	0.2903420 mol/mol	$17.0\cdot10^{-6}$ mol/mol	normal	$30\cdot10^{-6}$	$510\cdot10^{-12}$ g/g	0.0 %
R_{b10}	0.2924540 mol/mol	$46.5\cdot10^{-6}$ mol/mol	normal	$30\cdot10^{-6}$	$1.4\cdot10^{-9}$ g/g	0.0 %
R_{b2}	0.2864300 mol/mol	$41.5\cdot10^{-6}$ mol/mol	normal	$30\cdot10^{-6}$	$1.3\cdot10^{-9}$ g/g	0.0 %
R_{b3}	0.2911880 mol/mol	$19.5\cdot10^{-6}$ mol/mol	normal	$30\cdot10^{-6}$	$580\cdot10^{-12}$ g/g	0.0 %

Quantity	Value	Standard Uncertainty	Distribution	Sensitivity Coefficient	Uncertainty Contribution	Index
R_{b4}	0.2898800 mol/mol	$70.0\cdot10^{-6}$ mol/mol	normal	$30\cdot10^{-6}$	$2.1\cdot10^{-9}$ g/g	0.2 %
R_{b5}	0.2990830 mol/mol	$18.0\cdot10^{-6}$ mol/mol	normal	$29\cdot10^{-6}$	$520\cdot10^{-12}$ g/g	0.0 %
R_{b6}	0.2916920 mol/mol	$17.5\cdot10^{-6}$ mol/mol	normal	$30\cdot10^{-6}$	$520\cdot10^{-12}$ g/g	0.0 %
R_{b7}	0.2904600 mol/mol	$36.0\cdot10^{-6}$ mol/mol	normal	$30\cdot10^{-6}$	$1.1\cdot10^{-9}$ g/g	0.0 %
R_{b8}	0.2911480 mol/mol	$18.0\cdot10^{-6}$ mol/mol	normal	$30\cdot10^{-6}$	$540\cdot10^{-12}$ g/g	0.0 %
R_{b9}	0.2870160 mol/mol	$14.5\cdot10^{-6}$ mol/mol	normal	$30\cdot10^{-6}$	$440\cdot10^{-12}$ g/g	0.0 %
$Rd_{242/239}$	450.085 mol/mol	0.121 mol/mol		$-120\cdot10^{-12}$	$-15\cdot10^{-12}$ g/g	0.0 %
R_y	$75.700\cdot10^{-6}$ mol/mol	$400\cdot10^{-9}$ mol/mol	normal	$-300\cdot10^{-6}$	$-120\cdot10^{-12}$ g/g	0.0 %
δ_1	0.0	$70.0\cdot10^{-12}$	normal	24	$1.7\cdot10^{-9}$ g/g	0.0 %
δ_{10}	0.0	$70.0\cdot10^{-12}$	normal	24	$1.7\cdot10^{-9}$ g/g	0.0 %
δ_2	0.0	$70.0\cdot10^{-12}$	normal	24	$1.7\cdot10^{-9}$ g/g	0.0 %
δ_3	0.0	$70.0\cdot10^{-12}$	normal	24	$1.7\cdot10^{-9}$ g/g	0.0 %
δ_4	0.0	$70.0\cdot10^{-12}$	normal	24	$1.7\cdot10^{-9}$ g/g	0.0 %
δ_5	0.0	$70.0\cdot10^{-12}$	normal	24	$1.7\cdot10^{-9}$ g/g	0.0 %
δ_6	0.0	$70.0\cdot10^{-12}$	normal	24	$1.7\cdot10^{-9}$ g/g	0.0 %
δ_7	0.0	$70.0\cdot10^{-12}$	normal	24	$1.7\cdot10^{-9}$ g/g	0.0 %
δ_8	0.0	$70.0\cdot10^{-12}$	normal	24	$1.7\cdot10^{-9}$ g/g	0.0 %
δ_9	0.0	$70.0\cdot10^{-12}$	normal	24	$1.7\cdot10^{-9}$ g/g	0.0 %
Δt_1	0.457221081 a					
Δt_2	0.399726215 a					
Δt_{spike1}	3.7098 a					
Δt_{spike2}	3.7673 a					
τ_{239}	24100.0 a	11.0 a	normal	$390\cdot10^{-15}$	$4.3\cdot10^{-12}$ g/g	0.0 %
τ_{242}	$373.00\cdot10^{3}$ a	3000 a	normal	$180\cdot10^{-18}$	$550\cdot10^{-15}$ g/g	0.0 %
γ_{242Pu}	$86.7252\cdot10^{-6}$ g/g	$54.1\cdot10^{-9}$ g/g				

γ_{Pu}: mass content of total Pu in sample (here IRMM-049e)

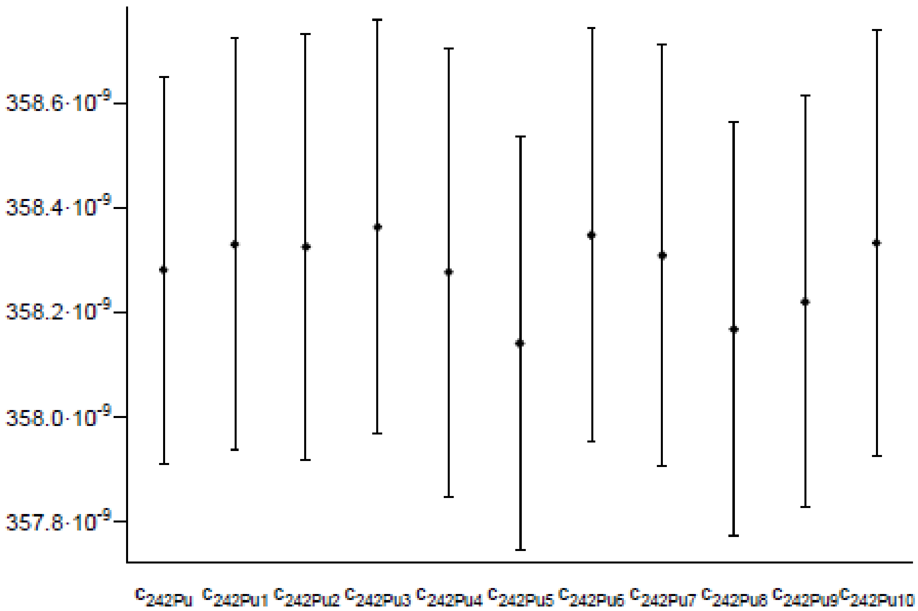
Quantity	Value	Standard Uncertainty	Distribution	Sensitivity Coefficient	Uncertainty Contribution	Index
$c_{HOM242Pu}$	0.0 mol/g	$66.3\cdot10^{-12}$ mol/g	normal	260	$17\cdot10^{-9}$ g/g	8.8 %
e	2.718281828459					
fd_{242}	0.9471745 mol/mol	$25.0\cdot10^{-6}$ mol/mol		$-97\cdot10^{-6}$	$-2.4\cdot10^{-9}$ g/g	0.2 %
K_{CRM}	1.000000	$500\cdot10^{-6}$	normal	$92\cdot10^{-6}$	$46\cdot10^{-9}$ g/g	64.1 %
K_{ext}	1.000000	$100\cdot10^{-6}$	normal	$92\cdot10^{-6}$	$9.2\cdot10^{-9}$ g/g	2.6 %
K_y	1.000000	$300\cdot10^{-6}$	normal	$92\cdot10^{-6}$	$27\cdot10^{-9}$ g/g	23.1 %
M_{239Pu}	239.05216360 g/mol	$1.90\cdot10^{-6}$ g/mol	normal	$-380\cdot10^{-9}$	$-730\cdot10^{-15}$ g/g	0.0 %
Md_{Pu}	241.9441670 g/mol	$57.2\cdot10^{-6}$ g/mol		$380\cdot10^{-9}$	$22\cdot10^{-12}$ g/g	0.0 %
m_{x1}	5.9990200 g	$45.0\cdot10^{-6}$ g	normal	$-1.5\cdot10^{-6}$	$-69\cdot10^{-12}$ g/g	0.0 %
m_{x10}	6.0334200 g	$50.0\cdot10^{-6}$ g	normal	$-1.5\cdot10^{-6}$	$-76\cdot10^{-12}$ g/g	0.0 %
m_{x2}	5.9332500 g	$40.0\cdot10^{-6}$ g	normal	$-1.5\cdot10^{-6}$	$-62\cdot10^{-12}$ g/g	0.0 %
m_{x3}	6.0183400 g	$40.0\cdot10^{-6}$ g	normal	$-1.5\cdot10^{-6}$	$-61\cdot10^{-12}$ g/g	0.0 %
m_{x4}	5.9734200 g	$60.0\cdot10^{-6}$ g	normal	$-1.5\cdot10^{-6}$	$-92\cdot10^{-12}$ g/g	0.0 %
m_{x5}	6.1806000 g	$40.0\cdot10^{-6}$ g	normal	$-1.5\cdot10^{-6}$	$-59\cdot10^{-12}$ g/g	0.0 %
m_{x6}	6.0092600 g	$60.0\cdot10^{-6}$ g	normal	$-1.5\cdot10^{-6}$	$-91\cdot10^{-12}$ g/g	0.0 %
m_{x7}	5.9984100 g	$40.0\cdot10^{-6}$ g	normal	$-1.5\cdot10^{-6}$	$-61\cdot10^{-12}$ g/g	0.0 %
m_{x8}	6.0221400 g	$55.0\cdot10^{-6}$ g	normal	$-1.5\cdot10^{-6}$	$-84\cdot10^{-12}$ g/g	0.0 %
m_{x9}	5.9300300 g	$50.0\cdot10^{-6}$ g	normal	$-1.5\cdot10^{-6}$	$-77\cdot10^{-12}$ g/g	0.0 %
n_{y1}	$1.7694\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 g/g	0.0 %
n_{y10}	$1.7667\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 g/g	0.0 %
n_{y2}	$1.7739\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 g/g	0.0 %
n_{y3}	$1.7701\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 g/g	0.0 %
n_{y4}	$1.7644\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 g/g	0.0 %
n_{y5}	$1.7687\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 g/g	0.0 %
n_{y6}	$1.7643\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 g/g	0.0 %
n_{y7}	$1.7684\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 g/g	0.0 %
n_{y8}	$1.7705\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 g/g	0.0 %
n_{y9}	$1.7688\cdot10^{-3}$ g	0.0 g	normal	0.0	0.0 g/g	0.0 %
R_{b1}	0.2903420 mol/mol	$17.0\cdot10^{-6}$ mol/mol	normal	$32\cdot10^{-6}$	$540\cdot10^{-12}$ g/g	0.0 %
R_{b10}	0.2924540 mol/mol	$46.5\cdot10^{-6}$ mol/mol	normal	$31\cdot10^{-6}$	$1.5\cdot10^{-9}$ g/g	0.0 %
R_{b2}	0.2864300 mol/mol	$41.5\cdot10^{-6}$ mol/mol	normal	$32\cdot10^{-6}$	$1.3\cdot10^{-9}$ g/g	0.0 %

Quantity	Value	Standard Uncertainty	Distribution	Sensitivity Coefficient	Uncertainty Contribution	Index
R_{b3}	0.2911880 mol/mol	$19.5\cdot10^{-6}$ mol/mol	normal	$31\cdot10^{-6}$	$610\cdot10^{-12}$ g/g	0.0 %
R_{b4}	0.2898800 mol/mol	$70.0\cdot10^{-6}$ mol/mol	normal	$32\cdot10^{-6}$	$2.2\cdot10^{-9}$ g/g	0.2 %
R_{b5}	0.2990830 mol/mol	$18.0\cdot10^{-6}$ mol/mol	normal	$31\cdot10^{-6}$	$550\cdot10^{-12}$ g/g	0.0 %
R_{b6}	0.2916920 mol/mol	$17.5\cdot10^{-6}$ mol/mol	normal	$31\cdot10^{-6}$	$550\cdot10^{-12}$ g/g	0.0 %
R_{b7}	0.2904600 mol/mol	$36.0\cdot10^{-6}$ mol/mol	normal	$32\cdot10^{-6}$	$1.1\cdot10^{-9}$ g/g	0.0 %
R_{b8}	0.2911480 mol/mol	$18.0\cdot10^{-6}$ mol/mol	normal	$31\cdot10^{-6}$	$570\cdot10^{-12}$ g/g	0.0 %
R_{b9}	0.2870160 mol/mol	$14.5\cdot10^{-6}$ mol/mol	normal	$32\cdot10^{-6}$	$460\cdot10^{-12}$ g/g	0.0 %
$Rd_{242/239}$	450.085 mol/mol	0.121 mol/mol		$-130\cdot10^{-12}$	$-16\cdot10^{-12}$ g/g	0.0 %
R_y	$75.700\cdot10^{-6}$ mol/mol	$400\cdot10^{-9}$ mol/mol	normal	$-310\cdot10^{-6}$	$-130\cdot10^{-12}$ g/g	0.0 %
δ_1	0.0	$70.0\cdot10^{-12}$	normal	26	$1.8\cdot10^{-9}$ g/g	0.0 %
δ_{10}	0.0	$70.0\cdot10^{-12}$	normal	26	$1.8\cdot10^{-9}$ g/g	0.0 %
δ_2	0.0	$70.0\cdot10^{-12}$	normal	26	$1.8\cdot10^{-9}$ g/g	0.0 %
δ_3	0.0	$70.0\cdot10^{-12}$	normal	26	$1.8\cdot10^{-9}$ g/g	0.0 %
δ_4	0.0	$70.0\cdot10^{-12}$	normal	26	$1.8\cdot10^{-9}$ g/g	0.0 %
δ_5	0.0	$70.0\cdot10^{-12}$	normal	26	$1.8\cdot10^{-9}$ g/g	0.0 %
δ_6	0.0	$70.0\cdot10^{-12}$	normal	26	$1.8\cdot10^{-9}$ g/g	0.0 %
δ_7	0.0	$70.0\cdot10^{-12}$	normal	26	$1.8\cdot10^{-9}$ g/g	0.0 %
δ_8	0.0	$70.0\cdot10^{-12}$	normal	26	$1.8\cdot10^{-9}$ g/g	0.0 %
δ_9	0.0	$70.0\cdot10^{-12}$	normal	26	$1.8\cdot10^{-9}$ g/g	0.0 %
Δt_1	0.457221081 a					
Δt_2	0.399726215 a					
Δt_{spike1}	3.7098 a					
Δt_{spike2}	3.7673 a					
τ_{239}	24100.0 a	11.0 a	normal	$410\cdot10^{-15}$	$4.5\cdot10^{-12}$ g/g	0.0 %
τ_{242}	$373.00\cdot10^{3}$ a	3000 a	normal	$200\cdot10^{-18}$	$590\cdot10^{-15}$ g/g	0.0 %
γ_{Pu}	$91.5186\cdot10^{-6}$ g/g	$57.2\cdot10^{-9}$ g/g				

Results:

Quantity	Value	Expanded Uncertainty	Coverage factor	Coverage
c_{242Pu}	$358.28\cdot10^{-9}$ mol/g	0.10 % (relative)	2.00	manual
c_{242Pu1}	$358.33\cdot10^{-9}$ mol/g	$390\cdot10^{-12}$ mol/g	2.00	manual
$c_{242Pu10}$	$358.33\cdot10^{-9}$ mol/g	$410\cdot10^{-12}$ mol/g	2.00	manual
c_{242Pu2}	$358.33\cdot10^{-9}$ mol/g	$410\cdot10^{-12}$ mol/g	2.00	manual
c_{242Pu3}	$358.36\cdot10^{-9}$ mol/g	$390\cdot10^{-12}$ mol/g	2.00	manual
c_{242Pu4}	$358.28\cdot10^{-9}$ mol/g	$430\cdot10^{-12}$ mol/g	2.00	manual
c_{242Pu5}	$358.14\cdot10^{-9}$ mol/g	$390\cdot10^{-12}$ mol/g	2.00	manual
c_{242Pu6}	$358.35\cdot10^{-9}$ mol/g	$390\cdot10^{-12}$ mol/g	2.00	95% (normal)
c_{242Pu7}	$358.31\cdot10^{-9}$ mol/g	$400\cdot10^{-12}$ mol/g	2.00	95% (normal)
c_{242Pu8}	$358.17\cdot10^{-9}$ mol/g	$390\cdot10^{-12}$ mol/g	2.00	95% (normal)
c_{242Pu9}	$358.22\cdot10^{-9}$ mol/g	$390\cdot10^{-12}$ mol/g	2.00	95% (normal)
$c_{242PuCRM}$	$358.28\cdot10^{-9}$ mol/g	0.12 % (relative)	2.00	95% (normal)
c_{Pu}	$378.26\cdot10^{-9}$ mol/g	0.12 % (relative)	2.00	manual
c_{x1}	$358.33\cdot10^{-9}$ mol/g	$370\cdot10^{-12}$ mol/g	2.00	95% (normal)
c_{x10}	$358.33\cdot10^{-9}$ mol/g	$380\cdot10^{-12}$ mol/g	2.00	95% (normal)
c_{x2}	$358.33\cdot10^{-9}$ mol/g	$380\cdot10^{-12}$ mol/g	2.00	95% (normal)
c_{x3}	$358.36\cdot10^{-9}$ mol/g	$370\cdot10^{-12}$ mol/g	2.00	95% (normal)
c_{x4}	$358.28\cdot10^{-9}$ mol/g	$400\cdot10^{-12}$ mol/g	2.00	95% (normal)
c_{x5}	$358.14\cdot10^{-9}$ mol/g	$370\cdot10^{-12}$ mol/g	2.00	95% (normal)
c_{x6}	$358.35\cdot10^{-9}$ mol/g	$370\cdot10^{-12}$ mol/g	2.00	95% (normal)
c_{x7}	$358.31\cdot10^{-9}$ mol/g	$380\cdot10^{-12}$ mol/g	2.00	95% (normal)
c_{x8}	$358.17\cdot10^{-9}$ mol/g	$370\cdot10^{-12}$ mol/g	2.00	95% (normal)
c_{x9}	$358.22\cdot10^{-9}$ mol/g	$370\cdot10^{-12}$ mol/g	2.00	95% (normal)
$R_x$dec1	450.08 mol/mol	0.52 mol/mol	2.00	95% (normal)
$R_x$dec2	450.08 mol/mol	0.52 mol/mol	2.00	95% (normal)
$R_y$dec1	$75.71\cdot10^{-6}$ mol/mol	$800\cdot10^{-9}$ mol/mol	2.00	95% (normal)
$R_y$dec2	$75.71\cdot10^{-6}$ mol/mol	$800\cdot10^{-9}$ mol/mol	2.00	95% (normal)
ε_1	$-50\cdot10^{-12}$	$140\cdot10^{-12}$	2.00	manual
ε_{10}	$-50\cdot10^{-12}$	$170\cdot10^{-12}$	2.00	manual
ε_2	$-40\cdot10^{-12}$	$160\cdot10^{-12}$	2.00	manual
ε_3	$-80\cdot10^{-12}$	$140\cdot10^{-12}$	2.00	manual
ε_4	0.0	$210\cdot10^{-12}$	2.00	manual
ε_5	$140\cdot10^{-12}$	$140\cdot10^{-12}$	2.00	manual

Quantity	Value	Expanded Uncertainty	Coverage factor	Coverage
ε_6	$-70\cdot10^{-12}$	$140\cdot10^{-12}$	2.00	manual
ε_7	$-30\cdot10^{-12}$	$160\cdot10^{-12}$	2.00	manual
ε_8	$110\cdot10^{-12}$	$140\cdot10^{-12}$	2.00	manual
ε_9	$60\cdot10^{-12}$	$140\cdot10^{-12}$	2.00	manual
γ_{242Pu}	$86.73\cdot10^{-6}$ g/g	0.12 % (relative)	2.00	manual
γ_{Pu}	$91.52\cdot10^{-6}$ g/g	0.12 % (relative)	2.00	95% (normal)
λ_{238}	$7.9000\cdot10^{-3}$ a^{-1}	$5.4\cdot10^{-6}$ a^{-1}	2.00	95% (normal)
λ_{239}	$28.761\cdot10^{-6}$ a^{-1}	$26\cdot10^{-9}$ a^{-1}	2.00	95% (normal)
λ_{240}	$105.65\cdot10^{-6}$ a^{-1}	$230\cdot10^{-9}$ a^{-1}	2.00	95% (normal)
λ_{241}	0.048387 a^{-1}	$81\cdot10^{-6}$ a^{-1}	2.00	95% (normal)
λ_{244}	$8.66\cdot10^{-9}$ a^{-1}	$190\cdot10^{-12}$ a^{-1}	2.00	95% (normal)



Annex 3.GUM file for Pu isotopic ratios determination in IRMM-049e

Certified IA for IRMM-049e- two series (after remeasurements)

FOR CERTIFICATE of amount ratios, moles and mass fractions

!RATIOS ARE CORRECTED FOR THE UNCERTAINTY FROM THE STANDARD IRMM-290b/A3 (K_{CRM}) AND FROM THE EXTERNAL CORRECTION (K_{ext})!

!! Note that for the final decayed ratios over 242, the uncertainty contribution from homogeneity is not taken into account!!

!!A consistency check was done, but no uncertainty for the delta values was applied (Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x)!!

ATTENTION! for the 238Pu/239Pu ratios the ratios from the TIMS turret (Cbis) were used since there was some contamination issue in 238U during the chemistry of TIMS turret E (re-measurement data)

Elements: Pu

Conversions and decay corrections for IA Pu ratios: 238/242, 239/242, 240/242, 241/242, 244/242 for IRMM-049e

DATA FOLDER:

Sample IRMM-049e to be certified Isotope amount ratios / NOTE THAT Superscript a, b, c, d refer respectively to different TIMS turrets: F, E, Cbis and D (turrets F and Cbis; E and D being associated for the IA ratios)

Date of measurements of IRMM-049e units: 7-17-24-28-37 (IDMS 18/07/2016) and 53-55-66-72-81 (IDMS 08/08/2016) IA analysis were carried out on units: 24- 28-53-66-81 on 22/09/2016 (first series) and 7-17-37-55-72 on 23/09/2016 (second series)

Date of reference: 1 January 2017

Mass metrology certificate : E3875

Report of analysis: E3593 (for IDMS and 238/242 IA analysis) and 3691 (for part of IA analysis)

Atomic mass: M. Wang et al: The AME2012 atomic mass evaluation (II). Tables, graphs and references

Half-lives: Monographie BIPM-5 Table of Radionuclides, Vol.5 and 241Pu half-life: Wellum et al. 2009 (JAAS)

Model Equation:

{in this GUMWB un-corrected ratios for 238Pu/239Pu, 240Pu/239Pu, 241Pu/239Pu, 242Pu/239Pu and 244Pu/239Pu are entered for IRMM-290-A3 (for k-factor determination) and for IRMM-049e. Those are measured in total evaporation (TE) without uncertainty on individual measurements.

Therefore they are type A, direct observation.}

{------------------------------ratios for IRMM-049e (1st series, IA analysis on 22/09/2016)--}

{decay correction for IRMM-290-A3}

$IRMM290A3cert_{242/239}=1/IRMM290A3cert_{239/242};$

$IRMM290A3certdec_{242/239}a=IRMM290A3cert_{242/239}\cdot(e\wedge(-\lambda_{242}\cdot\Delta t_{290A3}a)/e\wedge(-\lambda_{239}\cdot\Delta t_{290A3}a));$

$IRMM290A3certdec_{242/239}c=IRMM290A3cert_{242/239}\cdot(e\wedge(-\lambda_{242}\cdot\Delta t_{290A3}c)/e\wedge(-\lambda_{239}\cdot\Delta t_{290A3}c));$

{K-Factor calculations}

$K_{242/239}a=IRMM290A3certdec_{242/239}a/R_{242/239.IRMM290A3}a;$

$K_{242/239}c=IRMM290A3certdec_{242/239}c/R_{242/239.IRMM290A3}c;$

$K_{238/239}c=K_{242/239}c\wedge(-1/3);$

$K_{240/239}a=K_{242/239}a\wedge(1/3);$

$K_{241/239}a=K_{242/239}a$^(2/3);

$K_{244/239}a=K_{242/239}a$^(5/3);

{Corrections}

$Rcorr_{238/239}a=Runcorr_{238/239}c*K_{238/239}c$;

$Rcorr_{240/239}a=Runcorr_{240/239}a*K_{240/239}a$;

$Rcorr_{242/239}a=Runcorr_{242/239}a*K_{242/239}a$;

$Rcorr_{241/239}a=Runcorr_{241/239}a*K_{241/239}a$;

$Rcorr_{244/239}a=Runcorr_{244/239}a*K_{244/239}a$;

{-------------------------------conversion ratios for IRMM-049e (1st series, IA analysis on 22/09/2016)---------------------}

$Rcorr_{238/242}a=Rcorr_{238/239}a/Rcorr_{242/239}a$;

$Rcorr_{239/242}a=1/Rcorr_{242/239}a$;

$Rcorr_{240/242}a=Rcorr_{240/239}a/Rcorr_{242/239}a$;

$Rcorr_{241/242}a=Rcorr_{241/239}a/Rcorr_{242/239}a$;

$Rcorr_{244/242}a=Rcorr_{244/239}a/Rcorr_{242/239}a$;

{-------------------------------decayed isotopic ratios in the sample (1st series, IA analysis to reference date)-------------------------}

$Rd_{238/242}a = Rcorr_{238/242}a \cdot (e\text{^}(-\lambda_{238} \cdot \Delta t_{049e}c)/e\text{^}(-\lambda_{242} \cdot \Delta t_{049e}c))+\delta_1$;

$Rd_{239/242}a = Rcorr_{239/242}a \cdot (e\text{^}(-\lambda_{239} \cdot \Delta t_{049e}a)/e\text{^}(-\lambda_{242} \cdot \Delta t_{049e}a))+\delta_2$;

$Rd_{240/242}a = Rcorr_{240/242}a \cdot (e\text{^}(-\lambda_{240} \cdot \Delta t_{049e}a)/e\text{^}(-\lambda_{242} \cdot \Delta t_{049e}a))+\delta_3$;

$Rd_{241/242}a= Rcorr_{241/242}a \cdot (e\text{^}(-\lambda_{241} \cdot \Delta t_{049e}a)/e\text{^}(-\lambda_{242} \cdot \Delta t_{049e}a))+\delta_4$;

$Rd_{244/242}a = Rcorr_{244/242}a \cdot (e\text{^}(-\lambda_{244} \cdot \Delta t_{049e}a)/e\text{^}(-\lambda_{242} \cdot \Delta t_{049e}a))+\delta_5$;

{-------------------------------conversion decayed ratios for IRMM-049e (1st series, IA analysis to reference date)--------------------------}

$Rd_{238/239}a=Rd_{238/242}a/Rd_{239/242}a$;

$Rd_{240/239}a=Rd_{240/242}a/Rd_{239/242}a$;

$Rd_{241/239}a=Rd_{241/242}a/Rd_{239/242}a$;

$Rd_{242/239}a=1/Rd_{239/242}a$;

$Rd_{244/239}a=Rd_{244/242}a/Rd_{239/242}a$;

{-------------------------------ratios for IRMM-049e (2nd series, IA analysis on 23/09/2016)---}

{decay correction for IRMM-290-A3}

$IRMM290A3certdec_{242/239}b=IRMM290A3cert_{242/239} \cdot (e\text{^}(-\lambda_{242} \cdot \Delta t_{290A3}b)/e\text{^}(-\lambda_{239} \cdot \Delta t_{290A3}b))$;

$IRMM290A3certdec_{242/239}d=IRMM290A3cert_{242/239} \cdot (e\text{^}(-\lambda_{242} \cdot \Delta t_{290A3}d)/e\text{^}(-\lambda_{239} \cdot \Delta t_{290A3}d))$;

{K-Factor calculations}

$K_{242/239}b=IRMM290A3certdec_{242/239}b/R_{242/239.IRMM290A3}b$;

$K_{242/239}d=IRMM290A3certdec_{242/239}d/R_{242/239.IRMM290A3}d$;

$K_{238/239}d=K_{242/239}d$^(-1/3);

$K_{240/239}b=K_{242/239}b$^(1/3);

$K_{241/239}b=K_{242/239}b^{(2/3)}$;

$K_{244/239}b=K_{242/239}b^{(5/3)}$;

{Corrections}

$Rcorr_{238/239}b=Runcorr_{238/239}d*K_{238/239}d$;

$Rcorr_{240/239}b=Runcorr_{240/239}b*K_{240/239}b$;

$Rcorr_{242/239}b=Runcorr_{242/239}b*K_{242/239}b$;

$Rcorr_{241/239}b=Runcorr_{241/239}b*K_{241/239}b$;

$Rcorr_{244/239}b=Runcorr_{244/239}b*K_{244/239}b$;

{------------------------------conversion ratios for IRMM-049e (2nd series, IA analysis on 23/09/2016)---------------------}

$Rcorr_{238/242}b=Rcorr_{238/239}b/Rcorr_{242/239}b$;

$Rcorr_{239/242}b=1/Rcorr_{242/239}b$;

$Rcorr_{240/242}b=Rcorr_{240/239}b/Rcorr_{242/239}b$;

$Rcorr_{241/242}b=Rcorr_{241/239}b/Rcorr_{242/239}b$;

$Rcorr_{244/242}b=Rcorr_{244/239}b/Rcorr_{242/239}b$;

{------------------------------decayed isotopic ratios in the sample (2nd series, IA analysis to reference date)-----------------------}

$Rd_{238/242}b = Rcorr_{238/242}b \cdot (e^{\wedge}(-\lambda_{238} \cdot \Delta t_{049e}d)/e^{\wedge}(-\lambda_{242} \cdot \Delta t_{049e}d))+\delta_6$;

$Rd_{239/242}b = Rcorr_{239/242}b \cdot (e^{\wedge}(-\lambda_{239} \cdot \Delta t_{049e}b)/e^{\wedge}(-\lambda_{242} \cdot \Delta t_{049e}b))+\delta_7$;

$Rd_{240/242}b = Rcorr_{240/242}b \cdot (e^{\wedge}(-\lambda_{240} \cdot \Delta t_{049e}b)/e^{\wedge}(-\lambda_{242} \cdot \Delta t_{049e}b))+\delta_8$;

$Rd_{241/242}b= Rcorr_{241/242}b \cdot (e^{\wedge}(-\lambda_{241} \cdot \Delta t_{049e}b)/e^{\wedge}(-\lambda_{242} \cdot \Delta t_{049e}b))+\delta_9$;

$Rd_{244/242}b = Rcorr_{244/242}b \cdot (e^{\wedge}(-\lambda_{244} \cdot \Delta t_{049e}b)/e^{\wedge}(-\lambda_{242} \cdot \Delta t_{049e}b))+\delta_{10}$;

{------------------------------conversion decayed ratios for IRMM-049e (2nd series, IA analysis to reference date)-------------------}

$Rd_{238/239}b=Rd_{238/242}b/Rd_{239/242}b$;

$Rd_{240/239}b=Rd_{240/242}b/Rd_{239/242}b$;

$Rd_{241/239}b=Rd_{241/242}b/Rd_{239/242}b$;

$Rd_{242/239}b=1/Rd_{239/242}b$;

$Rd_{244/239}b=Rd_{244/242}b/Rd_{239/242}b$;

{------------------------------ratios for IRMM-049e (average of the two series a and b)--}

{-----average of decayed ratios over 239 to reference date-------}

$Rd_{238/239}= ((Rd_{238/239}a +Rd_{238/239}b)/2)*(K_{CRM}*K_{ext})^{(-1/3)}$;

$Rd_{240/239}= ((Rd_{240/239}a +Rd_{240/239}b)/2)*(K_{CRM}*K_{ext})^{(1/3)}$;

$Rd_{241/239}= ((Rd_{241/239}a +Rd_{241/239}b)/2)*(K_{CRM}*K_{ext})^{(2/3)}$;

$Rd_{242/239}= ((Rd_{242/239}a +Rd_{242/239}b)/2)*(K_{CRM}*K_{ext})^{(3/3)}$;

$Rd_{244/239}= ((Rd_{244/239}a +Rd_{244/239}b)/2)*(K_{CRM}*K_{ext})^{(5/3)}$;

{-----decayed ratios over 242 to reference date (for certification)------}

$Rd_{238/242}$=(($Rd_{238/242}a$ +$Rd_{238/242}b$)/2)*(K_{CRM}*K_{ext})^(-4/3);

$Rd_{239/242}$=(($Rd_{239/242}a$ +$Rd_{239/242}b$)/2)*(K_{CRM}*K_{ext})^(-3/3);

$Rd_{240/242}$=(($Rd_{240/242}a$ +$Rd_{240/242}b$)/2)*(K_{CRM}*K_{ext})^(-2/3);

$Rd_{241/242}$=(($Rd_{241/242}a$ +$Rd_{241/242}b$)/2)*(K_{CRM}*K_{ext})^(-1/3);

$Rd_{244/242}$=(($Rd_{244/242}a$ +$Rd_{244/242}b$)/2)*(K_{CRM}*K_{ext})^(2/3);

$\Sigma R_{Pu}=Rd_{238/242}+Rd_{239/242}+Rd_{240/242}+Rd_{241/242}+1+Rd_{244/242}$;

{---------------------- Consistency check (delta values)----------------------}

$\varepsilon_1 = Rd_{238/242} - Rd_{238/242}a$;

$\varepsilon_2 = Rd_{239/242} - Rd_{239/242}a$;

$\varepsilon_3 = Rd_{240/242} - Rd_{240/242}a$;

$\varepsilon_4 = Rd_{241/242} - Rd_{241/242}a$;

$\varepsilon_5 = Rd_{244/242} - Rd_{244/242}a$;

$\varepsilon_6 = Rd_{238/242} - Rd_{238/242}b$;

$\varepsilon_7 = Rd_{239/242} - Rd_{239/242}b$;

$\varepsilon_8 = Rd_{240/242} - Rd_{240/242}b$;

$\varepsilon_9 = Rd_{241/242} - Rd_{241/242}b$;

$\varepsilon_{10} = Rd_{244/242} - Rd_{244/242}b$;

{------------------Mole (f) and mass (w) fractions to reference date 01/01/2017------------}

$f_{238}=Rd_{238/242}/\Sigma R_{Pu}$;

$f_{239}=Rd_{239/242}/\Sigma R_{Pu}$;

$f_{240}=Rd_{240/242}/\Sigma R_{Pu}$;

$f_{241}=Rd_{241/242}/\Sigma R_{Pu}$;

$f_{242}=1/\Sigma R_{Pu}$;

$f_{244}=Rd_{244/242}/\Sigma R_{Pu}$;

$M_{Pu}=M_{238Pu}\cdot f_{238}+M_{239Pu}\cdot f_{239}+M_{240Pu}\cdot f_{240}+M_{241Pu}\cdot f_{241}+M_{242Pu}\cdot f_{242}+M_{244Pu}\cdot f_{244}$;

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

$w_{239}=f_{239}\cdot M_{239Pu}/M_{Pu}$;

$w_{240}=f_{240}\cdot M_{240Pu}/M_{Pu}$;

$w_{241}=f_{241}\cdot M_{241Pu}/M_{Pu}$;

$w_{242}=f_{242}\cdot M_{242Pu}/M_{Pu}$;

$w_{244}=f_{244}\cdot M_{244Pu}/M_{Pu}$;

{-----------parameters---------}

$ln_2 = ln(2)$;

$\lambda_{238} = ln_2 / \tau_{238}$;

$\lambda_{239} = ln_2 / \tau_{239}$;

$\lambda_{240} = ln_2 / \tau_{240}$;

$\lambda_{241} = ln_2 / \tau_{241}$;

$\lambda_{242} = ln_2 / \tau_{242}$;

$\lambda_{244} = ln_2 / \tau_{244}$;

List of Quantities:

Quantity	Unit	Definition
e		exponential
f_{238}	mol/mol	mole fraction of ^{238}Pu on reference date
f_{239}	mol/mol	mole fraction of ^{239}Pu on reference date
f_{240}	mol/mol	mole fraction of ^{240}Pu on reference date
f_{241}	mol/mol	mole fraction of ^{241}Pu on reference date
f_{242}	mol/mol	mole fraction of ^{242}Pu on reference date
f_{244}	mol/mol	mole fraction of ^{244}Pu on reference date
$IRMM290A3cert_{239/242}$	mol/mol	certified value for $^{239}Pu/^{242}Pu$ for IRMM-290-A3 (1 July 1986 and revised value)
$IRMM290A3cert_{242/239}$	mol/mol	certified value for $^{242}Pu/^{239}Pu$ for IRMM-290-A3 (1 July 1986 and revised value)
$IRMM290A3certdec_{242/239}a$	mol/mol	decayed certified value for $^{242}Pu/^{239}Pu$ for IRMM-290-A3 (IA analysis, turret F)
$IRMM290A3certdec_{242/239}b$	mol/mol	decayed certified value for $^{242}Pu/^{239}Pu$ for IRMM-290-A3 (IA analysis, turret E)
$IRMM290A3certdec_{242/239}c$	mol/mol	decayed certified value for $^{242}Pu/^{239}Pu$ for IRMM-290-A3 (IA analysis, turret Cbis)
$IRMM290A3certdec_{242/239}d$	mol/mol	decayed certified value for $^{242}Pu/^{239}Pu$ for IRMM-290-A3 (IA analysis, turret D)
$K_{238/239}c$		K-factor $^{238}Pu/^{239}Pu$ for turret Cbis
$K_{238/239}d$		K-factor $^{238}Pu/^{239}Pu$ for turret D
$K_{240/239}a$		K-factor $^{240}Pu/^{239}Pu$ for turret F
$K_{240/239}b$		K-factor $^{240}Pu/^{239}Pu$ for turret E
$K_{241/239}a$		K-factor $^{241}Pu/^{239}Pu$ for turret F
$K_{241/239}b$		K-factor $^{241}Pu/^{239}Pu$ for turret E
$K_{242/239}a$		K-factor $^{242}Pu/^{239}Pu$ for turret F
$K_{242/239}b$		K-factor $^{242}Pu/^{239}Pu$ for turret E
$K_{242/239}c$		K-factor $^{242}Pu/^{239}Pu$ for turret Cbis
$K_{242/239}d$		K-factor $^{242}Pu/^{239}Pu$ for turret D
$K_{244/239}a$		K-factor $^{244}Pu/^{239}Pu$ for turret F
$K_{244/239}b$		K-factor $^{244}Pu/^{239}Pu$ for turret E
K_{CRM}		correction factor for the uncertainty of the standard IRMM-290b/A3 used for mass fractionation
K_{ext}		correction factor for the uncertainty of the external correction during TIMS measurements
ln_2		constant
M_{238Pu}	g/mol	atomic mass for ^{238}Pu
M_{239Pu}	g/mol	atomic mass for ^{239}Pu

Quantity	Unit	Definition
M_{240Pu}	g/mol	atomic mass for ^{240}Pu
M_{241Pu}	g/mol	atomic mass for ^{241}Pu
M_{242Pu}	g/mol	atomic mass for ^{242}Pu
M_{244Pu}	g/mol	atomic mass for ^{244}Pu
M_{Pu}	g/mol	molar mass of Pu on DATE (reference date)
$R_{242/239.IRMM290A3}a$	mol/mol	measured values for $^{242}Pu/^{239}Pu$ for IRMM-290-A3 turret F
$R_{242/239.IRMM290A3}b$	mol/mol	measured values for $^{242}Pu/^{239}Pu$ for IRMM-290-A3 turret E
$R_{242/239.IRMM290A3}c$	mol/mol	measured values for $^{242}Pu/^{239}Pu$ for IRMM-290-A3 turret Cbis
$R_{242/239.IRMM290A3}d$	mol/mol	measured values for $^{242}Pu/^{239}Pu$ for IRMM-290-A3 turret D
$Rcorr_{238/239}a$	mol/mol	corrected value for $^{238}Pu/^{239}Pu$ for IRMM-049e turret F
$Rcorr_{238/239}b$	mol/mol	corrected value for $^{238}Pu/^{239}Pu$ for IRMM-049e turret E
$Rcorr_{238/242}a$	mol/mol	
$Rcorr_{238/242}b$	mol/mol	
$Rcorr_{239/242}a$	mol/mol	
$Rcorr_{239/242}b$	mol/mol	
$Rcorr_{240/239}a$	mol/mol	corrected value for $^{240}Pu/^{239}Pu$ for IRMM-049e turret F
$Rcorr_{240/239}b$	mol/mol	corrected value for $^{240}Pu/^{239}Pu$ for IRMM-049e turret E
$Rcorr_{240/242}a$	mol/mol	
$Rcorr_{240/242}b$	mol/mol	
$Rcorr_{241/239}a$	mol/mol	corrected value for $^{241}Pu/^{239}Pu$ for IRMM-049e turret F
$Rcorr_{241/239}b$	mol/mol	corrected value for $^{241}Pu/^{239}Pu$ for IRMM-049e turret E
$Rcorr_{241/242}a$	mol/mol	
$Rcorr_{241/242}b$	mol/mol	
$Rcorr_{242/239}a$	mol/mol	corrected value for $^{242}Pu/^{239}Pu$ for IRMM-049e turret F
$Rcorr_{242/239}b$	mol/mol	corrected value for $^{242}Pu/^{239}Pu$ for IRMM-049e turret E
$Rcorr_{244/239}a$	mol/mol	corrected value for $^{244}Pu/^{239}Pu$ for IRMM-049e turret F
$Rcorr_{244/239}b$	mol/mol	corrected value for $^{244}Pu/^{239}Pu$ for IRMM-049e turret E
$Rcorr_{244/242}a$	mol/mol	
$Rcorr_{244/242}b$	mol/mol	
$Rd_{238/239}$	mol/mol	average decayed ratios to reference date
$Rd_{238/239}a$	mol/mol	
$Rd_{238/239}b$	mol/mol	
$Rd_{238/242}$	mol/mol	average decayed ratios to reference date
$Rd_{238/242}a$	mol/mol	
$Rd_{238/242}b$	mol/mol	
$Rd_{239/242}$	mol/mol	average decayed ratios to reference date
$Rd_{239/242}a$	mol/mol	

Quantity	Unit	Definition
$Rd_{239/242}b$	mol/mol	
$Rd_{240/239}$	mol/mol	average decayed ratios to reference date
$Rd_{240/239}a$	mol/mol	
$Rd_{240/239}b$	mol/mol	
$Rd_{240/242}$	mol/mol	average decayed ratios to reference date
$Rd_{240/242}a$	mol/mol	
$Rd_{240/242}b$	mol/mol	
$Rd_{241/239}$	mol/mol	average decayed ratios to reference date
$Rd_{241/239}a$	mol/mol	
$Rd_{241/239}b$	mol/mol	
$Rd_{241/242}$	mol/mol	average decayed ratios to reference date
$Rd_{241/242}a$	mol/mol	
$Rd_{241/242}b$	mol/mol	
$Rd_{242/239}$	mol/mol	average decayed ratios to reference date
$Rd_{242/239}a$	mol/mol	
$Rd_{242/239}b$	mol/mol	
$Rd_{244/239}$	mol/mol	average decayed ratios to reference date
$Rd_{244/239}a$	mol/mol	
$Rd_{244/239}b$	mol/mol	
$Rd_{244/242}$	mol/mol	average decayed ratios to reference date
$Rd_{244/242}a$	mol/mol	
$Rd_{244/242}b$	mol/mol	
$Runcorr_{238/239}c$	mol/mol	measured value for $^{238}Pu/^{239}Pu$ for IRMM-049e turret Cbis
$Runcorr_{238/239}d$	mol/mol	measured value for $^{238}Pu/^{239}Pu$ for IRMM-049e turret D
$Runcorr_{240/239}a$	mol/mol	measured value for $^{240}Pu/^{239}Pu$ for IRMM-049e turret F
$Runcorr_{240/239}b$	mol/mol	measured value for $^{240}Pu/^{239}Pu$ for IRMM-049e turret E
$Runcorr_{241/239}a$	mol/mol	measured value for $^{241}Pu/^{239}Pu$ for IRMM-049e turret F
$Runcorr_{241/239}b$	mol/mol	measured value for $^{241}Pu/^{239}Pu$ for IRMM-049e turret E
$Runcorr_{242/239}a$	mol/mol	measured value for $^{242}Pu/^{239}Pu$ for IRMM-049e turret F
$Runcorr_{242/239}b$	mol/mol	measured value for $^{242}Pu/^{239}Pu$ for IRMM-049e turret E
$Runcorr_{244/239}a$	mol/mol	measured value for $^{244}Pu/^{239}Pu$ for IRMM-049e turret F
$Runcorr_{244/239}b$	mol/mol	measured value for $^{244}Pu/^{239}Pu$ for IRMM-049e turret E
w_{238}	g/g	mass fraction of ^{238}Pu
w_{239}	g/g	mass fraction of ^{239}Pu
w_{240}	g/g	mass fraction of ^{240}Pu
w_{241}	g/g	mass fraction of ^{241}Pu
w_{242}	g/g	mass fraction of ^{242}Pu
w_{244}	g/g	mass fraction of ^{244}Pu

Quantity	Unit	Definition
δ_1		delta values from the Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x
δ_{10}		delta values from the Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x
δ_2		delta values from the Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x
δ_3		delta values from the Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x
δ_4		delta values from the Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x
δ_5		delta values from the Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x
δ_6		delta values from the Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x
δ_7		delta values from the Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x
δ_8		delta values from the Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x
δ_9		delta values from the Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x
$\Delta t_{049e}a$	a	time difference between the IA analysis (1st series, IA analysis) and the reference date
$\Delta t_{049e}b$	a	time difference between the IA analysis (2nd series IA analysis) and the reference date
$\Delta t_{049e}c$	a	time difference between the IA analysis (238/242 analysed on turret Cbis) and the reference date
$\Delta t_{049e}d$	a	time difference between the IA analysis (238/242 analysed on turret D) and the reference date
$\Delta t_{290A3}a$	a	time difference between IA analysis date turret F and 1 July 1986 for IRMM-290-A3
$\Delta t_{290A3}b$	a	time difference between IA analysis date turret E and 1 July 1986 for IRMM-290-A3
$\Delta t_{290A3}c$	a	time difference between IA analysis date turret Cbis and 1 July 1986 for IRMM-290-A3
$\Delta t_{290A3}d$	a	time difference between IA analysis date turret D and 1 July 1986 for IRMM-290-A3
ε_1		Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x
ε_{10}		Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x
ε_2		Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x
ε_3		Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x

Quantity	Unit	Definition
ε_4		Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x
ε_5		Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x
ε_6		Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x
ε_7		Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x
ε_8		Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x
ε_9		Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x
λ_{238}	a^{-1}	decay constant ^{238}Pu
λ_{239}	a^{-1}	decay constant ^{239}Pu
λ_{240}	a^{-1}	decay constant ^{240}Pu
λ_{241}	a^{-1}	decay constant ^{241}Pu
λ_{242}	a^{-1}	decay constant ^{242}Pu
λ_{244}	a^{-1}	decay constant ^{244}Pu
ΣR_{Pu}		sum of the Pu amount ratios in IRMM-049e
τ_{238}	a	half life ^{238}Pu
τ_{239}	a	half life ^{239}Pu
τ_{240}	a	half life ^{240}Pu
τ_{241}	a	half life ^{241}Pu
τ_{242}	a	half life ^{242}Pu
τ_{244}	a	half life ^{244}Pu

e: Constant
Value: 2.7182818284590

IRMM290A3cert$_{239/242}$: Type B normal distribution
Value: 1.000700 mol/mol
Expanded Uncertainty: 0 mol/mol
Coverage Factor: 2

using NEW original certificate value for the IRMM-290b/A3 standard

K_{CRM}: Type B normal distribution
Value: 1
Expanded Uncertainty: 0.1 %
Coverage Factor: 2

uncertainty from standard IRMM-290b/A3 (see Excel file from PUTON measurements)

K_{ext}: Type B normal distribution
Value: 1
Expanded Uncertainty: 0.01 %
Coverage Factor: 1

uncertainty from external correction (see Excel file from PUTON measurements), 0.01% with k=1

M_{238Pu}: Type B normal distribution
Value: 238.0495601 g/mol
Expanded Uncertainty: 0.0000038 g/mol
Coverage Factor: 2

M. Wang et al: The AME2012 atomic mass evaluation (II)

M_{239Pu}: Type B normal distribution
Value: 239.0521636 g/mol
Expanded Uncertainty: 0.0000038 g/mol
Coverage Factor: 2

M. Wang et al: The AME2012 atomic mass evaluation (II)

M_{240Pu}: Type B normal distribution
Value: 240.0538138 g/mol
Expanded Uncertainty: 0.0000038 g/mol
Coverage Factor: 2

M. Wang et al: The AME2012 atomic mass evaluation (II)

M_{241Pu}: Type B normal distribution
Value: 241.0568517 g/mol
Expanded Uncertainty: 0.0000038 g/mol
Coverage Factor: 2

M. Wang et al: The AME2012 atomic mass evaluation (II)

M_{242Pu}: Type B normal distribution
Value: 242.0587428 g/mol
Expanded Uncertainty: 0.0000040 g/mol
Coverage Factor: 2

M. Wang et al: The AME2012 atomic mass evaluation (II)

M_{244Pu}: Type B normal distribution
Value: 244.064205 g/mol
Expanded Uncertainty: 0.000012 g/mol
Coverage Factor: 2

M. Wang et al: The AME2012 atomic mass evaluation (II)

$R_{242/239.IRMM290A3}$a: Type A
Method of observation: Direct
Number of observations: 6

No.	Observation
1	1.000349 mol/mol
2	1.000309 mol/mol
3	1.000255 mol/mol
4	1.000430 mol/mol
5	1.000473 mol/mol
6	1.000085 mol/mol

Arithmetic Mean: 1.0003168 mol/mol
Standard Deviation: $140 \cdot 10^{-6}$ mol/mol
Standard Uncertainty: $56.5 \cdot 10^{-6}$ mol/mol

Input data: P160922 Pu IRMM-049e turret F revision 8-CV-2.xls and file IA ratios IRMM049e turret F.xlsx

$R_{242/239.IRMM290A3}$b: Type A
Method of observation: Direct
Number of observations: 6

No.	Observation
1	1.000505 mol/mol
2	1.000295 mol/mol
3	1.000346 mol/mol
4	1.000439 mol/mol
5	1.000405 mol/mol
6	1.000377 mol/mol

Arithmetic Mean: 1.0003945 mol/mol
Standard Deviation: $73 \cdot 10^{-6}$ mol/mol
Standard Uncertainty: $29.9 \cdot 10^{-6}$ mol/mol

Input data: P160923 Pu IRMM-049e turret E revision 8-CV-2.xls and file IA ratios IRMM049e turret E.xlsx

$R_{242/239.IRMM290A3}$c: Type A
Method of observation: Direct
Number of observations: 6

No.	Observation
1	1.000256 mol/mol
2	1.000383 mol/mol
3	1.000252 mol/mol
4	1.000274 mol/mol
5	1.000312 mol/mol
6	1.000282 mol/mol

Arithmetic Mean: 1.0002932 mol/mol
Standard Deviation: $49 \cdot 10^{-6}$ mol/mol
Standard Uncertainty: $20.0 \cdot 10^{-6}$ mol/mol

P160811 IRMM-049e IA TurretCbis-CV-2.xls

$R_{242/239.IRMM290A3}$d: Type A
Method of observation: Direct
Number of observations: 6

No.	Observation
1	1.000307 mol/mol
2	1.000264 mol/mol
3	1.000327 mol/mol
4	1.000328 mol/mol
5	1.000439 mol/mol
6	1.000354 mol/mol

Arithmetic Mean: 1.0003365 mol/mol
Standard Deviation: $58 \cdot 10^{-6}$ mol/mol
Standard Uncertainty: $23.9 \cdot 10^{-6}$ mol/mol

P160824 IRMM-049e IA TurretD-CV-2.xls

$Runcorr_{238/239}c$: Type A
Method of observation: Direct
Number of observations: 5

No.	Observation
1	2.289727 mol/mol
2	2.29096 mol/mol
3	2.284311 mol/mol
4	2.285898 mol/mol
5	2.287628 mol/mol

Arithmetic Mean: 2.28770 mol/mol
Standard Deviation: $2.7 \cdot 10^{-3}$ mol/mol
Standard Uncertainty: $1.21 \cdot 10^{-3}$ mol/mol

P160811 IRMM-049e IA TurretCbis-CV-2.xls

$Runcorr_{238/239}d$: Type A
Method of observation: Direct
Number of observations: 5

No.	Observation
1	2.284819 mol/mol
2	2.288480 mol/mol
3	2.285946 mol/mol
4	2.284589 mol/mol
5	2.287365 mol/mol

Arithmetic Mean: 2.286240 mol/mol
Standard Deviation: $1.7 \cdot 10^{-3}$ mol/mol
Standard Uncertainty: $745 \cdot 10^{-6}$ mol/mol

P160824 IRMM-049e IA TurretD-CV-2.xls

$Runcorr_{240/239}a$: Type A
Method of observation: Direct
Number of observations: 5

No.	Observation
1	20.71925863 mol/mol
2	20.70407178 mol/mol
3	20.72002153 mol/mol
4	20.7303091 mol/mol
5	20.73514257 mol/mol

Arithmetic Mean: 20.72176 mol/mol
Standard Deviation: 0.012 mol/mol
Standard Uncertainty: $5.36 \cdot 10^{-3}$ mol/mol

P160922 Pu IRMM-049e turret F revision 8-CV-2.xls

$Runcorr_{240/239}b$: Type A
Method of observation: Direct
Number of observations: 5

No.	Observation
1	20.7245114 mol/mol
2	20.69075853 mol/mol
3	20.71028598 mol/mol
4	20.73181049 mol/mol
5	20.74110903 mol/mol

Arithmetic Mean: 20.71970 mol/mol
Standard Deviation: 0.020 mol/mol
Standard Uncertainty: $8.82 \cdot 10^{-3}$ mol/mol

P160923 Pu IRMM-049e turret E revision 8-CV-2.xls

$Runcorr_{241/239}a$: Type A
Method of observation: Direct
Number of observations: 5

No.	Observation
1	1.001645811 mol/mol
2	0.999552143 mol/mol
3	1.001561518 mol/mol
4	0.999082693 mol/mol
5	1.001299674 mol/mol

Arithmetic Mean: 1.000628 mol/mol
Standard Deviation: $1.2 \cdot 10^{-3}$ mol/mol
Standard Uncertainty: $543 \cdot 10^{-6}$ mol/mol

P160922 Pu IRMM-049e turret F revision 8-CV-2.xls

$Runcorr_{241/239}b$: Type A
Method of observation: Direct
Number of observations: 5

No.	Observation
1	1.00232628 mol/mol
2	0.999856919 mol/mol
3	0.998745215 mol/mol
4	1.00044654 mol/mol
5	0.999535753 mol/mol

Arithmetic Mean: 1.000182 mol/mol
Standard Deviation: $1.3 \cdot 10^{-3}$ mol/mol
Standard Uncertainty: $602 \cdot 10^{-6}$ mol/mol

P160923 Pu IRMM-049e turret E revision 8-CV-2.xls

$Runcorr_{242/239}a$: Type A
Method of observation: Direct
Number of observations: 5

No.	Observation
1	450.1325773 mol/mol
2	449.7138467 mol/mol
3	450.1294353 mol/mol
4	450.4538139 mol/mol
5	450.5989363 mol/mol

Arithmetic Mean: 450.206 mol/mol
Standard Deviation: 0.34 mol/mol
Standard Uncertainty: 0.153 mol/mol

P160922 Pu IRMM-049e turret F revision 8-CV-2.xls

$Runcorr_{242/239}b$: Type A
Method of observation: Direct
Number of observations: 5

No.	Observation
1	450.2999413 mol/mol
2	449.5581791 mol/mol
3	449.9827379 mol/mol
4	450.4130491 mol/mol
5	450.6240231 mol/mol

Arithmetic Mean: 450.176 mol/mol
Standard Deviation: 0.42 mol/mol
Standard Uncertainty: 0.186 mol/mol

P160923 Pu IRMM-049e turret E revision 8-CV-2.xls

$Runcorr_{244/239}a$: Type A
Method of observation: Direct
Number of observations: 5

No.	Observation
1	0.116035883 mol/mol
2	0.115362967 mol/mol
3	0.115715251 mol/mol
4	0.115801675 mol/mol
5	0.116523615 mol/mol

Arithmetic Mean: 0.115888 mol/mol
Standard Deviation: $430 \cdot 10^{-6}$ mol/mol
Standard Uncertainty: $192 \cdot 10^{-6}$ mol/mol

P160922 Pu IRMM-049e turret F revision 8-CV-2.xls

$Runcorr_{244/239}b$: Type A
Method of observation: Direct
Number of observations: 5

No.	Observation
1	0.115995026 mol/mol
2	0.115890543 mol/mol
3	0.116346344 mol/mol
4	0.115839154 mol/mol
5	0.116622325 mol/mol

Arithmetic Mean: 0.116139 mol/mol
Standard Deviation: $340 \cdot 10^{-6}$ mol/mol
Standard Uncertainty: $150 \cdot 10^{-6}$ mol/mol

P160923 Pu IRMM-049e turret E revision 8-CV-2.xls

δ_1: Type B normal distribution
Value: 0
Expanded Uncertainty: 0
Coverage Factor: 2

Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x

δ_{10}: Type B normal distribution
Value: 0
Expanded Uncertainty: 0
Coverage Factor: 2

Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x

δ_2: Type B normal distribution
Value: 0
Expanded Uncertainty: 0
Coverage Factor: 2

Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x

δ_3: Type B normal distribution
Value: 0
Expanded Uncertainty: 0
Coverage Factor: 2

Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x

δ_4: Type B normal distribution
Value: 0
Expanded Uncertainty: 0
Coverage Factor: 2

Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x

δ_5: Type B normal distribution
Value: 0
Expanded Uncertainty: 0
Coverage Factor: 2

Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x

δ_6: Type B normal distribution
Value: 0
Expanded Uncertainty: 0
Coverage Factor: 2

Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x

δ_7: Type B normal distribution
Value: 0
Expanded Uncertainty: 0
Coverage Factor: 2

Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x

δ_8: Type B normal distribution
Value: 0
Expanded Uncertainty: 0
Coverage Factor: 2

Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x

δ_9: Type B normal distribution
Value: 0
Expanded Uncertainty: 0
Coverage Factor: 2

Consistency check Kessel et al. (2008) Accred Qual Assur. DOI 10.1007/s00769-008-0382-x

$\Delta t_{049e}a$: Constant
Value: 0.27652293 a

time difference between IA analysis 22/09/2016 and reference date 01/01/2017 (101 days)

$\Delta t_{049e}b$: Constant
Value: 0.273785079 a

time difference between IA analysis 23/09/2016 and reference date 01/01/2017 (100 days)

$\Delta t_{049e}c$: Constant
Value: 0.3915 a

time difference between 11/08/2016 and 01/01/2017 = 0.3915 a

$\Delta t_{049e}d$: Constant
Value: 0.3559 a

time difference between 24/08/2016 and 01/01/2017 = 0.3559 a

$\Delta t_{290A3}a$: Constant
Value: 30.2286 a

from P160922 Pu IRMM-049e turret F revision 8-CV.xls

$\Delta t_{290A3}b$: Constant
Value: 30.2313 a

from P160923 Pu IRMM-049e turret E revision 8-CV.xls

$\Delta t_{290A3}c$: Constant
Value: 30.1136 a

as from P160811 IRMM-049e IA TurretCbis-CV-2.xls

$\Delta t_{290A3}d$: Constant
Value: 30.1492 a

as from P160824 IRMM-049e IA TurretD-CV-2.xls

τ_{238}: Type B normal distribution
Value: 87.74 a
Expanded Uncertainty: 0.03 a
Coverage Factor: 1

Monographie BIPM-5 Table of Radionuclides, Vol.5

τ_{239}: Type B normal distribution
Value: 24100 a
Expanded Uncertainty: 11 a
Coverage Factor: 1

Monographie BIPM-5 Table of Radionuclides, Vol.5

τ_{240}: Type B normal distribution
Value: 6561 a
Expanded Uncertainty: 7 a
Coverage Factor: 1

Monographie BIPM-5 Table of Radionuclides, Vol.5

τ_{241}: Type B normal distribution
Value: 14.325 a
Expanded Uncertainty: 0.024 a
Coverage Factor: 2

Wellum et al. (2009) JAAS

τ_{242}: Type B normal distribution
Value: 373000 a
Expanded Uncertainty: 3000 a
Coverage Factor: 1

Monographie BIPM-5 Table of Radionuclides, Vol.5

τ_{244}: Type B normal distribution
Value: $8 \cdot 10^7$ a
Expanded Uncertainty: $0.09 \cdot 10^7$ a
Coverage Factor: 1

Monographie BIPM-5 Table of Radionuclides, Vol.5

Interim Results:

Quantity	Value	Standard Uncertainty
$IRMM290A3certdec_{242/239}a$	1.000113491 mol/mol	$601 \cdot 10^{-9}$ mol/mol
$IRMM290A3certdec_{242/239}b$	1.000113564 mol/mol	$601 \cdot 10^{-9}$ mol/mol
$IRMM290A3certdec_{242/239}c$	1.000110397 mol/mol	$599 \cdot 10^{-9}$ mol/mol
$IRMM290A3certdec_{242/239}d$	1.000111355 mol/mol	$600 \cdot 10^{-9}$ mol/mol
$K_{238/239}c$	1.00006091	$6.67 \cdot 10^{-6}$
$K_{238/239}d$	1.00007503	$7.96 \cdot 10^{-6}$
$K_{240/239}a$	0.9999322	$18.8 \cdot 10^{-6}$
$K_{240/239}b$	0.99990638	$9.97 \cdot 10^{-6}$
$K_{241/239}a$	0.9998645	$37.7 \cdot 10^{-6}$
$K_{241/239}b$	0.9998128	$19.9 \cdot 10^{-6}$
$K_{242/239}a$	0.9997967	$56.5 \cdot 10^{-6}$
$K_{242/239}b$	0.9997192	$29.9 \cdot 10^{-6}$
$K_{242/239}c$	0.9998173	$20.0 \cdot 10^{-6}$
$K_{242/239}d$	0.9997749	$23.9 \cdot 10^{-6}$
$K_{244/239}a$	0.9996612	$94.2 \cdot 10^{-6}$
$K_{244/239}b$	0.9995320	$49.8 \cdot 10^{-6}$
$Rcorr_{238/239}a$	2.28784 mol/mol	$1.21 \cdot 10^{-3}$ mol/mol
$Rcorr_{238/239}b$	2.286411 mol/mol	$746 \cdot 10^{-6}$ mol/mol
$Rcorr_{240/239}a$	20.72036 mol/mol	$5.37 \cdot 10^{-3}$ mol/mol
$Rcorr_{240/239}b$	20.71776 mol/mol	$8.82 \cdot 10^{-3}$ mol/mol
$Rcorr_{241/239}a$	1.000493 mol/mol	$545 \cdot 10^{-6}$ mol/mol
$Rcorr_{241/239}b$	0.999995 mol/mol	$603 \cdot 10^{-6}$ mol/mol
$Rcorr_{242/239}a$	450.114 mol/mol	0.155 mol/mol
$Rcorr_{242/239}b$	450.049 mol/mol	0.186 mol/mol
$Rcorr_{244/239}a$	0.115849 mol/mol	$192 \cdot 10^{-6}$ mol/mol
$Rcorr_{244/239}b$	0.116084 mol/mol	$150 \cdot 10^{-6}$ mol/mol
$Rd_{238/239}a$	2.28080 mol/mol	$1.21 \cdot 10^{-3}$ mol/mol
$Rd_{238/239}b$	2.280010 mol/mol	$744 \cdot 10^{-6}$ mol/mol
$Rd_{240/239}a$	20.71992 mol/mol	$5.37 \cdot 10^{-3}$ mol/mol
$Rd_{240/239}b$	20.71732 mol/mol	$8.82 \cdot 10^{-3}$ mol/mol
$Rd_{241/239}a$	0.987203 mol/mol	$537 \cdot 10^{-6}$ mol/mol
$Rd_{241/239}b$	0.986842 mol/mol	$595 \cdot 10^{-6}$ mol/mol
$Rd_{242/239}a$	450.118 mol/mol	0.155 mol/mol
$Rd_{242/239}b$	450.052 mol/mol	0.186 mol/mol
$Rd_{244/239}a$	0.115850 mol/mol	$192 \cdot 10^{-6}$ mol/mol
$Rd_{244/239}b$	0.116085 mol/mol	$150 \cdot 10^{-6}$ mol/mol

Quantity	Value	Standard Uncertainty
λ_{238}	$7.90001 \cdot 10^{-3}$ a^{-1}	$2.70 \cdot 10^{-6}$ a^{-1}
λ_{239}	$28.7613 \cdot 10^{-6}$ a^{-1}	$13.1 \cdot 10^{-9}$ a^{-1}
λ_{240}	$105.647 \cdot 10^{-6}$ a^{-1}	$113 \cdot 10^{-9}$ a^{-1}
λ_{241}	0.0483872 a^{-1}	$40.5 \cdot 10^{-6}$ a^{-1}
λ_{242}	$1.8583 \cdot 10^{-6}$ a^{-1}	$14.9 \cdot 10^{-9}$ a^{-1}
λ_{244}	$8.6643 \cdot 10^{-9}$ a^{-1}	$97.5 \cdot 10^{-12}$ a^{-1}
ΣR_{Pu}	1.0557717	$27.9 \cdot 10^{-6}$

Uncertainty Budgets:

$Rd_{239/242}$: **average decayed ratios to reference date**

Quantity	Value	Standard Uncertainty	Distribution	Sensitivity Coefficient	Uncertainty Contribution	Index
e	2.718281828459					
$IRMM290A3cert_{239/242}$	1.0007 mol/mol	0.0 mol/mol	normal	0.0	0.0 mol/mol	0.0 %
K_{CRM}	1.000000	$500 \cdot 10^{-6}$	normal	$-2.2 \cdot 10^{-3}$	$-1.1 \cdot 10^{-6}$ mol/mol	75.2 %
K_{ext}	1.000000	$100 \cdot 10^{-6}$	normal	$-2.2 \cdot 10^{-3}$	$-220 \cdot 10^{-9}$ mol/mol	3.0 %
$R_{242/239.IRMM290A3}a$	1.0003168 mol/mol	$56.5 \cdot 10^{-6}$ mol/mol	normal	$1.1 \cdot 10^{-3}$	$63 \cdot 10^{-9}$ mol/mol	0.2 %
$R_{242/239.IRMM290A3}b$	1.0003945 mol/mol	$29.9 \cdot 10^{-6}$ mol/mol	normal	$1.1 \cdot 10^{-3}$	$33 \cdot 10^{-9}$ mol/mol	0.0 %
$Runcorr_{242/239}a$	450.206 mol/mol	0.153 mol/mol	normal	$-2.5 \cdot 10^{-6}$	$-380 \cdot 10^{-9}$ mol/mol	8.7 %
$Runcorr_{242/239}b$	450.176 mol/mol	0.186 mol/mol	normal	$-2.5 \cdot 10^{-6}$	$-460 \cdot 10^{-9}$ mol/mol	12.8 %
δ_2	0.0	0.0	normal	0.0	0.0 mol/mol	0.0 %
δ_7	0.0	0.0	normal	0.0	0.0 mol/mol	0.0 %
$\Delta t_{049e}a$	0.27652293 a					
$\Delta t_{049e}b$	0.273785079 a					
$\Delta t_{290A3}a$	30.2286 a					
$\Delta t_{290A3}b$	30.2313 a					
τ_{239}	24100.0 a	11.0 a	normal	$81 \cdot 10^{-12}$	$890 \cdot 10^{-12}$ mol/mol	0.0 %
τ_{242}	$373.00 \cdot 10^{3}$ a	3000 a	normal	$-340 \cdot 10^{-15}$	$-1.0 \cdot 10^{-9}$ mol/mol	0.0 %
$Rd_{239/242}$	$2.22180 \cdot 10^{-3}$ mol/mol	$1.28 \cdot 10^{-6}$ mol/mol				

Results:

Quantity	Value	Expanded Uncertainty	Coverage factor	Coverage
f_{238}	$4.7990 \cdot 10^{-3}$ mol/mol	$7.4 \cdot 10^{-6}$ mol/mol	2.00	95% (normal)
f_{239}	$2.1044 \cdot 10^{-3}$ mol/mol	$2.3 \cdot 10^{-6}$ mol/mol	2.00	95% (normal)
f_{240}	0.043601 mol/mol	$41 \cdot 10^{-6}$ mol/mol	2.00	95% (normal)
f_{241}	$2.0771 \cdot 10^{-3}$ mol/mol	$2.1 \cdot 10^{-6}$ mol/mol	2.00	95% (normal)
f_{242}	0.947174 mol/mol	$50 \cdot 10^{-6}$ mol/mol	2.00	95% (normal)
f_{244}	$244.05 \cdot 10^{-6}$ mol/mol	$560 \cdot 10^{-9}$ mol/mol	2.00	95% (normal)
M_{Pu}	241.94417 g/mol	$110 \cdot 10^{-6}$ g/mol	2.00	manual
$Rcorr_{238/242}a$	$5.0828 \cdot 10^{-3}$ mol/mol	$6.4 \cdot 10^{-6}$ mol/mol	2.00	95% (normal)
$Rcorr_{238/242}b$	$5.0804 \cdot 10^{-3}$ mol/mol	$5.4 \cdot 10^{-6}$ mol/mol	2.00	95% (normal)
$Rcorr_{239/242}a$	$2.2217 \cdot 10^{-3}$ mol/mol	$1.5 \cdot 10^{-6}$ mol/mol	2.00	95% (normal)
$Rcorr_{239/242}b$	$2.2220 \cdot 10^{-3}$ mol/mol	$1.8 \cdot 10^{-6}$ mol/mol	2.00	95% (normal)
$Rcorr_{240/242}a$	0.046034 mol/mol	$39 \cdot 10^{-6}$ mol/mol	2.00	95% (normal)
$Rcorr_{240/242}b$	0.046034 mol/mol	$55 \cdot 10^{-6}$ mol/mol	2.00	95% (normal)
$Rcorr_{241/242}a$	$2.2228 \cdot 10^{-3}$ mol/mol	$2.8 \cdot 10^{-6}$ mol/mol	2.00	95% (normal)
$Rcorr_{241/242}b$	$2.2220 \cdot 10^{-3}$ mol/mol	$3.2 \cdot 10^{-6}$ mol/mol	2.00	95% (normal)
$Rcorr_{244/242}a$	$257.38 \cdot 10^{-6}$ mol/mol	$870 \cdot 10^{-9}$ mol/mol	2.00	95% (normal)
$Rcorr_{244/242}b$	$257.94 \cdot 10^{-6}$ mol/mol	$700 \cdot 10^{-9}$ mol/mol	2.00	95% (normal)
$Rd_{238/239}$	2.2804 mol/mol	$1.6 \cdot 10^{-3}$ mol/mol	2.00	95% (normal)
$Rd_{238/242}$	$5.0666 \cdot 10^{-3}$ mol/mol	$8.1 \cdot 10^{-6}$ mol/mol	2.00	95% (normal)
$Rd_{238/242}a$	$5.0671 \cdot 10^{-3}$ mol/mol	$6.4 \cdot 10^{-6}$ mol/mol	2.00	95% (normal)
$Rd_{238/242}b$	$5.0661 \cdot 10^{-3}$ mol/mol	$5.3 \cdot 10^{-6}$ mol/mol	2.00	95% (normal)
$Rd_{239/242}$	$2.2218 \cdot 10^{-3}$ mol/mol	$2.6 \cdot 10^{-6}$ mol/mol	2.00	95% (normal)
$Rd_{239/242}a$	$2.2216 \cdot 10^{-3}$ mol/mol	$1.5 \cdot 10^{-6}$ mol/mol	2.00	95% (normal)

Quantity	Value	Expanded Uncertainty	Coverage factor	Coverage
$Rd_{239/242}b$	$2.2220\cdot10^{-3}$ mol/mol	$1.8\cdot10^{-6}$ mol/mol	2.00	95% (normal)
$Rd_{240/239}$	20.719 mol/mol	0.012 mol/mol	2.00	95% (normal)
$Rd_{240/242}$	0.046033 mol/mol	$46\cdot10^{-6}$ mol/mol	2.00	95% (normal)
$Rd_{240/242}a$	0.046032 mol/mol	$39\cdot10^{-6}$ mol/mol	2.00	95% (normal)
$Rd_{240/242}b$	0.046033 mol/mol	$55\cdot10^{-6}$ mol/mol	2.00	95% (normal)
$Rd_{241/239}$	0.9870 mol/mol	$1.0\cdot10^{-3}$ mol/mol	2.00	95% (normal)
$Rd_{241/242}$	$2.1930\cdot10^{-3}$ mol/mol	$2.3\cdot10^{-6}$ mol/mol	2.00	95% (normal)
$Rd_{241/242}a$	$2.1932\cdot10^{-3}$ mol/mol	$2.8\cdot10^{-6}$ mol/mol	2.00	95% (normal)
$Rd_{241/242}b$	$2.1927\cdot10^{-3}$ mol/mol	$3.2\cdot10^{-6}$ mol/mol	2.00	95% (normal)
$Rd_{242/239}$	450.09 mol/mol	0.52 mol/mol	2.00	95% (normal)
$Rd_{244/239}$	0.11597 mol/mol	$310\cdot10^{-6}$ mol/mol	2.00	95% (normal)
$Rd_{244/242}$	$257.66\cdot10^{-6}$ mol/mol	$590\cdot10^{-9}$ mol/mol	2.00	95% (normal)
$Rd_{244/242}a$	$257.38\cdot10^{-6}$ mol/mol	$870\cdot10^{-9}$ mol/mol	2.00	95% (normal)
$Rd_{244/242}b$	$257.94\cdot10^{-6}$ mol/mol	$700\cdot10^{-9}$ mol/mol	2.00	95% (normal)
w_{238}	$4.7217\cdot10^{-3}$ g/g	$7.3\cdot10^{-6}$ g/g	2.00	95% (normal)
w_{239}	$2.0793\cdot10^{-3}$ g/g	$2.3\cdot10^{-6}$ g/g	2.00	95% (normal)
w_{240}	0.043260 g/g	$41\cdot10^{-6}$ g/g	2.00	95% (normal)
w_{241}	$2.0695\cdot10^{-3}$ g/g	$2.1\cdot10^{-6}$ g/g	2.00	95% (normal)
w_{242}	0.947623 g/g	$50\cdot10^{-6}$ g/g	2.00	95% (normal)
w_{244}	$246.18\cdot10^{-6}$ g/g	$560\cdot10^{-9}$ g/g	2.00	95% (normal)
ε_1	$-500\cdot10^{-9}$	$8.1\cdot10^{-6}$	2.00	manual
ε_{10}	$-280\cdot10^{-9}$	$590\cdot10^{-9}$	2.00	manual
ε_2	$200\cdot10^{-9}$	$2.6\cdot10^{-6}$	2.00	manual
ε_3	0.0	$46\cdot10^{-6}$	2.00	manual
ε_4	$-200\cdot10^{-9}$	$2.3\cdot10^{-6}$	2.00	manual
ε_5	$280\cdot10^{-9}$	$590\cdot10^{-9}$	2.00	manual
ε_6	$500\cdot10^{-9}$	$8.1\cdot10^{-6}$	2.00	manual
ε_7	$-200\cdot10^{-9}$	$2.6\cdot10^{-6}$	2.00	manual
ε_8	0.0	$46\cdot10^{-6}$	2.00	manual
ε_9	$200\cdot10^{-9}$	$2.3\cdot10^{-6}$	2.00	manual

Annex 4. Input data for the ANOVA of the homogeneity assessment (single values, means and standard deviations of the three replicates in analytical sequence order) in analytical sequence order for the ^{242}Pu amount content and the Pu isotope amount ratios of IRMM-049e

^{242}Pu amount content [mol/g]					
Unit	**Replicate 1**	**Replicate 2**	**Replicate 3**	**Sample Mean**	***s***
37	$3.5822\cdot10^{-7}$	$3.5816\cdot10^{-7}$	$3.5818\cdot10^{-7}$	$3.5819\cdot10^{-7}$	$3.10\cdot10^{-11}$
17	$3.5843\cdot10^{-7}$	$3.5833\cdot10^{-7}$	/	$3.5838\cdot10^{-7}$	$7.10\cdot10^{-11}$
28	$3.5847\cdot10^{-7}$	$3.5818\cdot10^{-7}$	$3.5829\cdot10^{-7}$	$3.5831\cdot10^{-7}$	$1.46\cdot10^{-10}$
7	$3.5835\cdot10^{-7}$	$3.5841\cdot10^{-7}$	$3.5837\cdot10^{-7}$	$3.5838\cdot10^{-7}$	$3.10\cdot10^{-11}$
24	$3.5844\cdot10^{-7}$	$3.5837\cdot10^{-7}$	$3.5842\cdot10^{-7}$	$3.5841\cdot10^{-7}$	$3.60\cdot10^{-11}$
66	$3.5819\cdot10^{-7}$	$3.5822\cdot10^{-7}$	$3.5824\cdot10^{-7}$	$3.5822\cdot10^{-7}$	$2.50\cdot10^{-11}$
81	$3.5848\cdot10^{-7}$	$3.5833\cdot10^{-7}$	$3.5831\cdot10^{-7}$	$3.5837\cdot10^{-7}$	$9.30\cdot10^{-11}$
53	$3.5838\cdot10^{-7}$	$3.5838\cdot10^{-7}$	$3.5842\cdot10^{-7}$	$3.5839\cdot10^{-7}$	$2.30\cdot10^{-11}$
72	$3.5829\cdot10^{-7}$	$3.5828\cdot10^{-7}$	$3.5826\cdot10^{-7}$	$3.5828\cdot10^{-7}$	$1.50\cdot10^{-11}$
55	$3.5828\cdot10^{-7}$	$3.5837\cdot10^{-7}$	$3.5842\cdot10^{-7}$	$3.5836\cdot10^{-7}$	$7.10\cdot10^{-11}$

$n(^{238}\text{Pu})/n(^{242}\text{Pu})$ [mol/mol]					
Unit	**Replicate 1**	**Replicate 2**	**Replicate 3**	**Sample Mean**	***s***
37	$5.0644\cdot10^{-3}$	$5.0612\cdot10^{-3}$	$5.0651\cdot10^{-3}$	$5.0636\cdot10^{-3}$	$2.1\cdot10^{-6}$
17	$5.0676\cdot10^{-3}$	$5.0652\cdot10^{-3}$	$5.0687\cdot10^{-3}$	$5.0672\cdot10^{-3}$	$1.8\cdot10^{-6}$
28	$5.0643\cdot10^{-3}$	$5.0658\cdot10^{-3}$	$5.0676\cdot10^{-3}$	$5.0659\cdot10^{-3}$	$1.7\cdot10^{-6}$
7	$5.0636\cdot10^{-3}$	$5.0629\cdot10^{-3}$	$5.0699\cdot10^{-3}$	$5.0655\cdot10^{-3}$	$3.9\cdot10^{-6}$
24	$5.0536\cdot10^{-3}$	$5.0686\cdot10^{-3}$	$5.0640\cdot10^{-3}$	$5.0621\cdot10^{-3}$	$7.7\cdot10^{-6}$
66	$5.0652\cdot10^{-3}$	$5.0616\cdot10^{-3}$	$5.0633\cdot10^{-3}$	$5.0634\cdot10^{-3}$	$1.8\cdot10^{-6}$
81	$5.0619\cdot10^{-3}$	$5.0650\cdot10^{-3}$	$5.0668\cdot10^{-3}$	$5.0646\cdot10^{-3}$	$2.5\cdot10^{-6}$
53	$5.0620\cdot10^{-3}$	$5.0604\cdot10^{-3}$	$5.0643\cdot10^{-3}$	$5.0622\cdot10^{-3}$	$2.0\cdot10^{-6}$
72	$5.0604\cdot10^{-3}$	$5.0634\cdot10^{-3}$	$5.0603\cdot10^{-3}$	$5.0614\cdot10^{-3}$	$1.8\cdot10^{-6}$
55	$5.0644\cdot10^{-3}$	$5.0620\cdot10^{-3}$	$5.0581\cdot10^{-3}$	$5.0615\cdot10^{-3}$	$3.2\cdot10^{-6}$

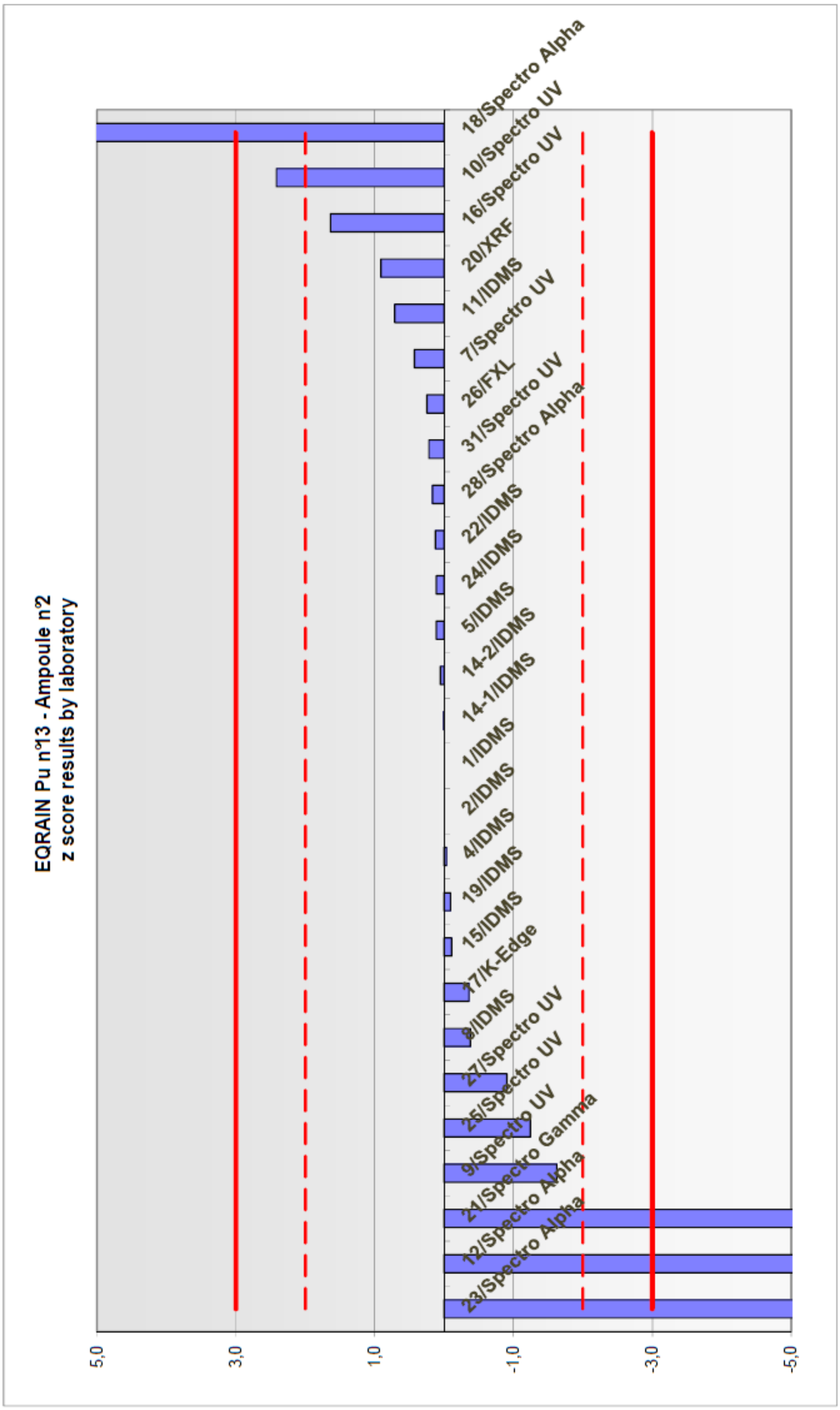
$n(^{239}Pu)/n(^{242}Pu)$ [mol/mol]					
Unit	**Replicate 1**	**Replicate 2**	**Replicate 3**	**Sample Mean**	**s**
28	$2.214\cdot10^{-3}$	$2.229\cdot10^{-3}$	$2.223\cdot10^{-3}$	$2.222\cdot10^{-3}$	$7.533\cdot10^{-6}$
53	$2.222\cdot10^{-3}$	$2.227\cdot10^{-3}$	$2.224\cdot10^{-3}$	$2.224\cdot10^{-3}$	$2.363\cdot10^{-6}$
81	$2.216\cdot10^{-3}$	$2.228\cdot10^{-3}$	$2.222\cdot10^{-3}$	$2.222\cdot10^{-3}$	$6.165\cdot10^{-6}$
24	$2.220\cdot10^{-3}$	$2.220\cdot10^{-3}$	$2.221\cdot10^{-3}$	$2.220\cdot10^{-3}$	$6.070\cdot10^{-7}$
66	$2.221\cdot10^{-3}$	$2.216\cdot10^{-3}$	$2.223\cdot10^{-3}$	$2.220\cdot10^{-3}$	$3.360\cdot10^{-6}$
17	$2.220\cdot10^{-3}$	$2.223\cdot10^{-3}$	$2.222\cdot10^{-3}$	$2.221\cdot10^{-3}$	$1.457\cdot10^{-6}$
55	$2.226\cdot10^{-3}$	$2.223\cdot10^{-3}$	$2.226\cdot10^{-3}$	$2.225\cdot10^{-3}$	$1.823\cdot10^{-6}$
7	$2.220\cdot10^{-3}$	$2.227\cdot10^{-3}$	$2.222\cdot10^{-3}$	$2.223\cdot10^{-3}$	$3.353\cdot10^{-6}$
72	$2.218\cdot10^{-3}$	$2.224\cdot10^{-3}$	$2.220\cdot10^{-3}$	$2.221\cdot10^{-3}$	$2.653\cdot10^{-6}$
37	$2.218\cdot10^{-3}$	$2.219\cdot10^{-3}$	$2.222\cdot10^{-3}$	$2.220\cdot10^{-3}$	$2.126\cdot10^{-6}$

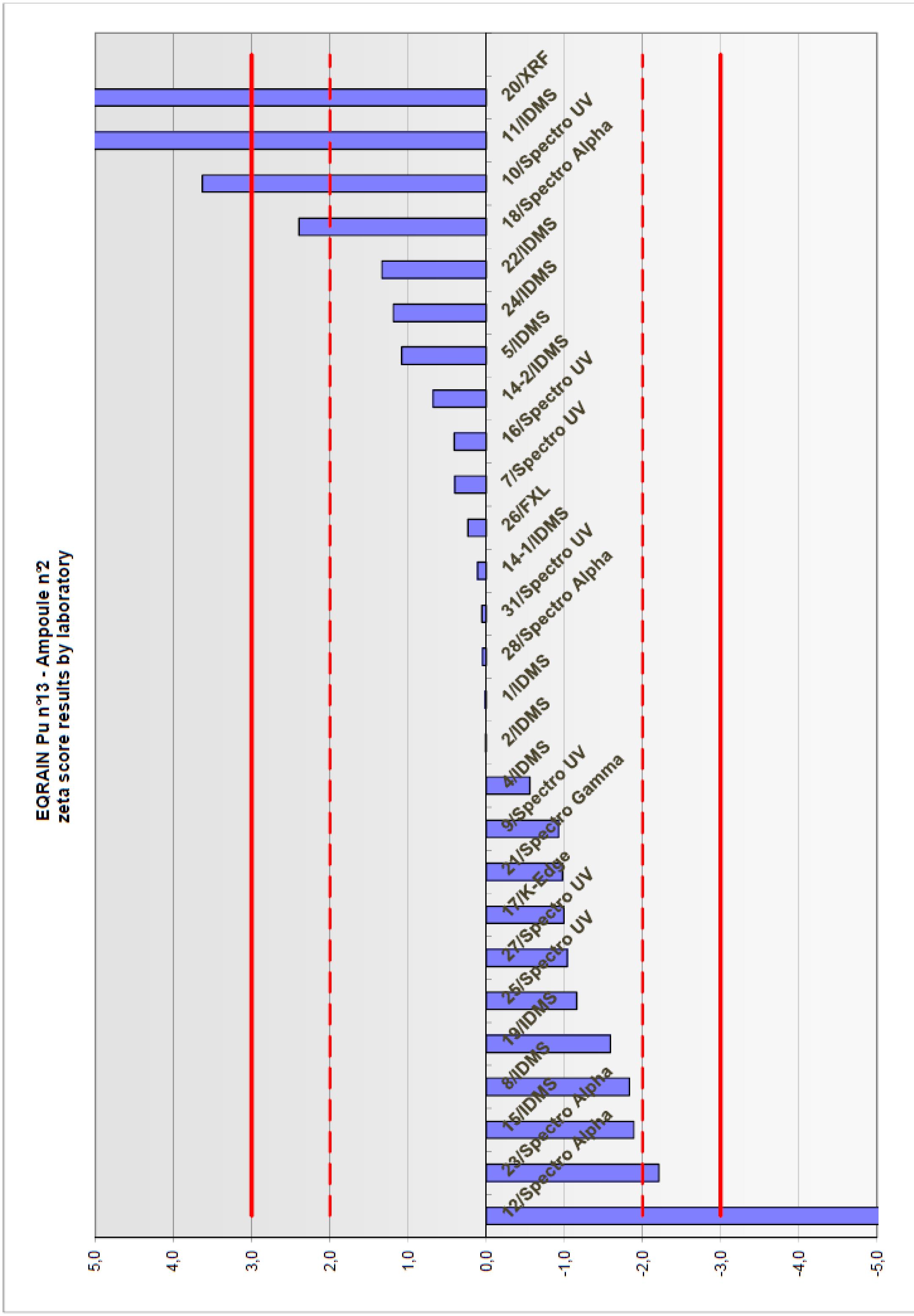
$n(^{240}Pu)/n(^{242}Pu)$ [mol/mol]					
Unit	**Replicate 1**	**Replicate 2**	**Replicate 3**	**Sample Mean**	**s**
28	$4.6029\cdot10^{-2}$	$4.6030\cdot10^{-2}$	$4.6043\cdot10^{-2}$	$4.6034\cdot10^{-2}$	$8.00\cdot10^{-6}$
53	$4.6044\cdot10^{-2}$	$4.6039\cdot10^{-2}$	$4.6046\cdot10^{-2}$	$4.6043\cdot10^{-2}$	$4.00\cdot10^{-6}$
81	$4.6031\cdot10^{-2}$	$4.6046\cdot10^{-2}$	$4.6031\cdot10^{-2}$	$4.6036\cdot10^{-2}$	$9.00\cdot10^{-6}$
24	$4.6031\cdot10^{-2}$	$4.6029\cdot10^{-2}$	$4.6018\cdot10^{-2}$	$4.6026\cdot10^{-2}$	$7.00\cdot10^{-6}$
66	$4.6015\cdot10^{-2}$	$4.6010\cdot10^{-2}$	$4.6040\cdot10^{-2}$	$4.6022\cdot10^{-2}$	$1.60\cdot10^{-6}$
17	$4.6037\cdot10^{-2}$	$4.6033\cdot10^{-2}$	$4.6023\cdot10^{-2}$	$4.6031\cdot10^{-2}$	$7.00\cdot10^{-6}$
55	$4.6029\cdot10^{-2}$	$4.6037\cdot10^{-2}$	$4.6030\cdot10^{-2}$	$4.6032\cdot10^{-2}$	$4.00\cdot10^{-6}$
7	$4.6034\cdot10^{-2}$	$4.6028\cdot10^{-2}$	$4.6034\cdot10^{-2}$	$4.6032\cdot10^{-2}$	$3.00\cdot10^{-6}$
72	$4.6044\cdot10^{-2}$	$4.6031\cdot10^{-2}$	$4.6032\cdot10^{-2}$	$4.6036\cdot10^{-2}$	$7.00\cdot10^{-6}$
37	$4.6034\cdot10^{-2}$	$4.6031\cdot10^{-2}$	$4.6039\cdot10^{-2}$	$4.6035\cdot10^{-2}$	$4.00\cdot10^{-6}$

$n(^{241}Pu)/n(^{242}Pu)$ [mol/mol]					
Unit	**Replicate 1**	**Replicate 2**	**Replicate 3**	**Sample Mean**	**s**
28	$2.1892 \cdot 10^{-3}$	$2.1994 \cdot 10^{-3}$	$2.1989 \cdot 10^{-3}$	$2.1958 \cdot 10^{-3}$	$5.80 \cdot 10^{-6}$
53	$2.1954 \cdot 10^{-3}$	$2.1905 \cdot 10^{-3}$	$2.1939 \cdot 10^{-3}$	$2.1932 \cdot 10^{-3}$	$2.50 \cdot 10^{-6}$
81	$2.1902 \cdot 10^{-3}$	$2.2038 \cdot 10^{-3}$	$2.1929 \cdot 10^{-3}$	$2.1956 \cdot 10^{-3}$	$7.20 \cdot 10^{-6}$
24	$2.1844 \cdot 10^{-3}$	$2.1920 \cdot 10^{-3}$	$2.1894 \cdot 10^{-3}$	$2.1886 \cdot 10^{-3}$	$3.90 \cdot 10^{-6}$
66	$2.1902 \cdot 10^{-3}$	$2.1930 \cdot 10^{-3}$	$2.1952 \cdot 10^{-3}$	$2.1928 \cdot 10^{-3}$	$2.50 \cdot 10^{-6}$
17	$2.1958 \cdot 10^{-3}$	$2.1970 \cdot 10^{-3}$	$2.1976 \cdot 10^{-3}$	$2.1968 \cdot 10^{-3}$	$9.00 \cdot 10^{-7}$
55	$2.1955 \cdot 10^{-3}$	$2.1916 \cdot 10^{-3}$	$2.1980 \cdot 10^{-3}$	$2.1950 \cdot 10^{-3}$	$3.20 \cdot 10^{-6}$
7	$2.1923 \cdot 10^{-3}$	$2.1915 \cdot 10^{-3}$	$2.1877 \cdot 10^{-3}$	$2.1905 \cdot 10^{-3}$	$2.40 \cdot 10^{-6}$
72	$2.1965 \cdot 10^{-3}$	$2.1915 \cdot 10^{-3}$	$2.1884 \cdot 10^{-3}$	$2.1921 \cdot 10^{-3}$	$4.10 \cdot 10^{-6}$
37	$2.1878 \cdot 10^{-3}$	$2.1929 \cdot 10^{-3}$	$2.1867 \cdot 10^{-3}$	$2.1891 \cdot 10^{-3}$	$3.30 \cdot 10^{-6}$

$n(^{244}Pu)/n(^{242}Pu)$ [mol/mol]					
Unit	**Replicate 1**	**Replicate 2**	**Replicate 3**	**Sample Mean**	**s**
28	$2.5787 \cdot 10^{-4}$	$2.5717 \cdot 10^{-4}$	$2.5820 \cdot 10^{-4}$	$2.5770 \cdot 10^{-4}$	$5.00 \cdot 10^{-7}$
53	$2.5656 \cdot 10^{-4}$	$2.5680 \cdot 10^{-4}$	$2.5611 \cdot 10^{-4}$	$2.5650 \cdot 10^{-4}$	$3.00 \cdot 10^{-7}$
81	$2.5610 \cdot 10^{-4}$	$2.5808 \cdot 10^{-4}$	$2.5694 \cdot 10^{-4}$	$2.5700 \cdot 10^{-4}$	$1.00 \cdot 10^{-6}$
24	$2.5556 \cdot 10^{-4}$	$2.5662 \cdot 10^{-4}$	$2.5895 \cdot 10^{-4}$	$2.5700 \cdot 10^{-4}$	$1.70 \cdot 10^{-6}$
66	$2.5915 \cdot 10^{-4}$	$2.5927 \cdot 10^{-4}$	$2.5726 \cdot 10^{-4}$	$2.5860 \cdot 10^{-4}$	$1.10 \cdot 10^{-6}$
17	$2.5729 \cdot 10^{-4}$	$2.5760 \cdot 10^{-4}$	$2.5776 \cdot 10^{-4}$	$2.5750 \cdot 10^{-4}$	$2.00 \cdot 10^{-7}$
55	$2.5842 \cdot 10^{-4}$	$2.5756 \cdot 10^{-4}$	$2.5725 \cdot 10^{-4}$	$2.5770 \cdot 10^{-4}$	$6.00 \cdot 10^{-7}$
7	$2.6036 \cdot 10^{-4}$	$2.5682 \cdot 10^{-4}$	$2.5835 \cdot 10^{-4}$	$2.5850 \cdot 10^{-4}$	$1.80 \cdot 10^{-6}$
72	$2.5698 \cdot 10^{-4}$	$2.5699 \cdot 10^{-4}$	$2.5744 \cdot 10^{-4}$	$2.5710 \cdot 10^{-4}$	$3.00 \cdot 10^{-7}$
37	$2.5767 \cdot 10^{-4}$	$2.6123 \cdot 10^{-4}$	$2.5736 \cdot 10^{-4}$	$2.5880 \cdot 10^{-4}$	$2.20 \cdot 10^{-6}$

Annex 5. Evaluation of laboratory performances by means of z- and ζ- scores during the plutonium EQRAIN-13 circuit





The laboratory performance of JRC.G.2-Geel (laboratory 19) was evaluated against the EQRAIN-13 reference value established at CEA/CETAMA and was found satisfactory, as expressed by means of the respective z- and ζ- scores.

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