



^{243}Am certified reference material for mass spectrometry

Rožle Jakopič¹ · Adelheid Fankhauser² · Yetunde Aregbe¹ · Stephan Richter¹ · Marielle Crozet³ ·
Christophe Maillard³ · Cédric Rivier³ · Danièle Roudil³ · Maria Marouli¹ · Faidra Tzika¹ · Timotheos Altzitzoglou¹ ·
Stefaan Pommé¹

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Abstract

The Joint Research Centre, in cooperation with the Commissariat à l’Energie Atomique et aux Energies Alternatives, produced a novel ^{243}Am spike reference material for mass spectrometry. Americium solution with an isotopic composition of 88% ^{243}Am and 12% ^{241}Am was used as the source for the preparation of the spike material. The certified value of 5.696 (11) nmol g⁻¹ for the amount content of ^{243}Am and 0.136138 (54) for the $n(^{241}\text{Am})/n(^{243}\text{Am})$ amount ratio were assigned. The assigned values from mass spectrometry were confirmed by alpha-particle spectrometry, alpha-particle counting at a defined solid angle, and high-resolution gamma-ray spectrometry. Furthermore, an external validation of the certified values was obtained from the results of an interlaboratory comparison exercise, using this americium reference solution as the test sample.

Keywords ^{243}Am · Nuclear safeguards · Nuclear forensics · Spike · Isotope dilution mass spectrometry (IDMS) · Certified reference material (CRM)

Introduction

Americium (Am) is a non-naturally occurring element, produced by neutron capture in nuclear reactions. Among various Am radioactive isotopes, ^{241}Am and ^{243}Am are the two important isotopes with half-lives of 432.6 (1.2) [1] and 7345 (28) years ($k=2$) [2]. ^{241}Am and other isotopes of Am are usually determined by alpha-particle and gamma-ray spectrometry [3, 4]. However, mass-spectrometric techniques have become increasingly important in nuclear safeguards and nuclear security, particularly when Am measurement results with lower uncertainty are needed. To achieve this, suitable certified reference materials (CRMs) are indispensable for calibration of mass spectrometers, validation of methods and in quality control.

Until today, no Am reference materials for mass spectrometry certified for the isotopic composition or amount content have been available on the market. Ideally, spike CRMs are produced gravimetrically from well-characterised high-purity metals and compounds such as purified oxides. Unlike for uranium and plutonium, americium base materials are scarce or difficult to obtain. Consequently, this has resulted in rare application of mass-spectrometric techniques for determination of Am. The need for an isotopic Am CRM has been expressed by the International Atomic Energy Agency (IAEA) and at the 2016 Nuclear Security Summit [5, 6]. Such material can be applied in nuclear forensics to determine accurately the model age of plutonium (Pu) materials, i.e. the time elapsed since its last chemical purification, via $^{241}\text{Am}/^{241}\text{Pu}$ chronometry [7–11], in radiation waste management for characterisation of irradiated nuclear fuel/waste for long-term disposal at geological repositories [12], and in nuclear decay data application for the determination of nuclear half-lives.

To fill this gap, the Joint Research Centre of the European Commission (EC-JRC) in Geel and the Commissariat à l’Energie Atomique et aux Energies Alternatives in Marcoule jointly produced and certified an ^{243}Am spike reference material. The Am starting material, enriched in

✉ Rožle Jakopič
rozle.jakopic@ec.europa.eu

¹ European Commission, Joint Research Centre (JRC), Geel, Belgium
² Paul Scherrer Institute (PSI), 5232 Villigen PSI, Switzerland
³ Commissariat à l’Energie Atomique et aux Energies Alternatives (CEA), DES, ISEC, DMRC, University of Montpellier, Montpellier, France

^{243}Am (88%), was provided by the Atalante Analysis Laboratory (CEA/L2AT, Marcoule). The isotopic composition of the Am source is different from that found in the fuel cycle or in purification processes, making it suitable as the source for this spike material and for the quantification of Am by mass spectrometry. Nearly 600 units of this reference material were produced, with a mass fraction of Am of about $1.5 \mu\text{g g}^{-1}$ solution. The CEA and the JRC-Geel each received half of the total units produced. The material was certified for the Am content by isotope dilution mass spectrometry (IDMS), using an ^{241}Am spike produced in-house from ^{241}Pu material, and for the isotope amount ratios (e.g. isotopic abundance) by thermal ionisation mass spectrometry (TIMS). The preparation and certification of this material were done in compliance with ISO 17034 and the ISO Guide 35 [13, 14]. Radiometric techniques were used as independent confirmation measurements. In parallel to the certification process and prior to release of the material's certificate, an interlaboratory comparison (ILC) was organised by the CEA's Commission d'ETablissement des Méthodes d'Analyse (CETAMA) using this reference material as the test sample [15].

In this paper, the preparation and the characterisation of this ^{243}Am spike CRM (IRMM-0243/STAM) will be presented. All uncertainty values in this paper are expanded uncertainties with a coverage factor $k = 2$. They are given in parentheses.

Methodology

The solution was characterised for the amount content of Am (e.g. ^{243}Am , ^{241}Am and total Am) using isotope dilution mass spectrometry (IDMS) as a primary method of measurement [16, 17]. In IDMS, the amount of an element (or isotope) in the sample is determined on the basis of additions of known amounts of the same element (called a spike) whose isotopic composition differs from that of the unknown sample. By measuring the change in isotopic composition of the blend (sample-spike mixture) by isotope mass spectrometry, the unknown amount of the element (or isotope) can be derived.

In absence of a suitable, well-characterised ^{241}Am spike, an alternative approach had to be chosen. The ^{241}Am spike was produced from highly enriched ^{241}Pu (99.3%) base material available at JRC-Geel. Two strategies for the characterisation of the ^{241}Am in-house spike were considered. The first strategy is based on separating the ingrown ^{241}Am from the ^{241}Pu material since its last purification and, subsequently, characterising the ^{241}Am amount content and the isotopic composition by means of mass spectrometry or radiometric techniques. This strategy, however, would require an ^{243}Am spike, which was

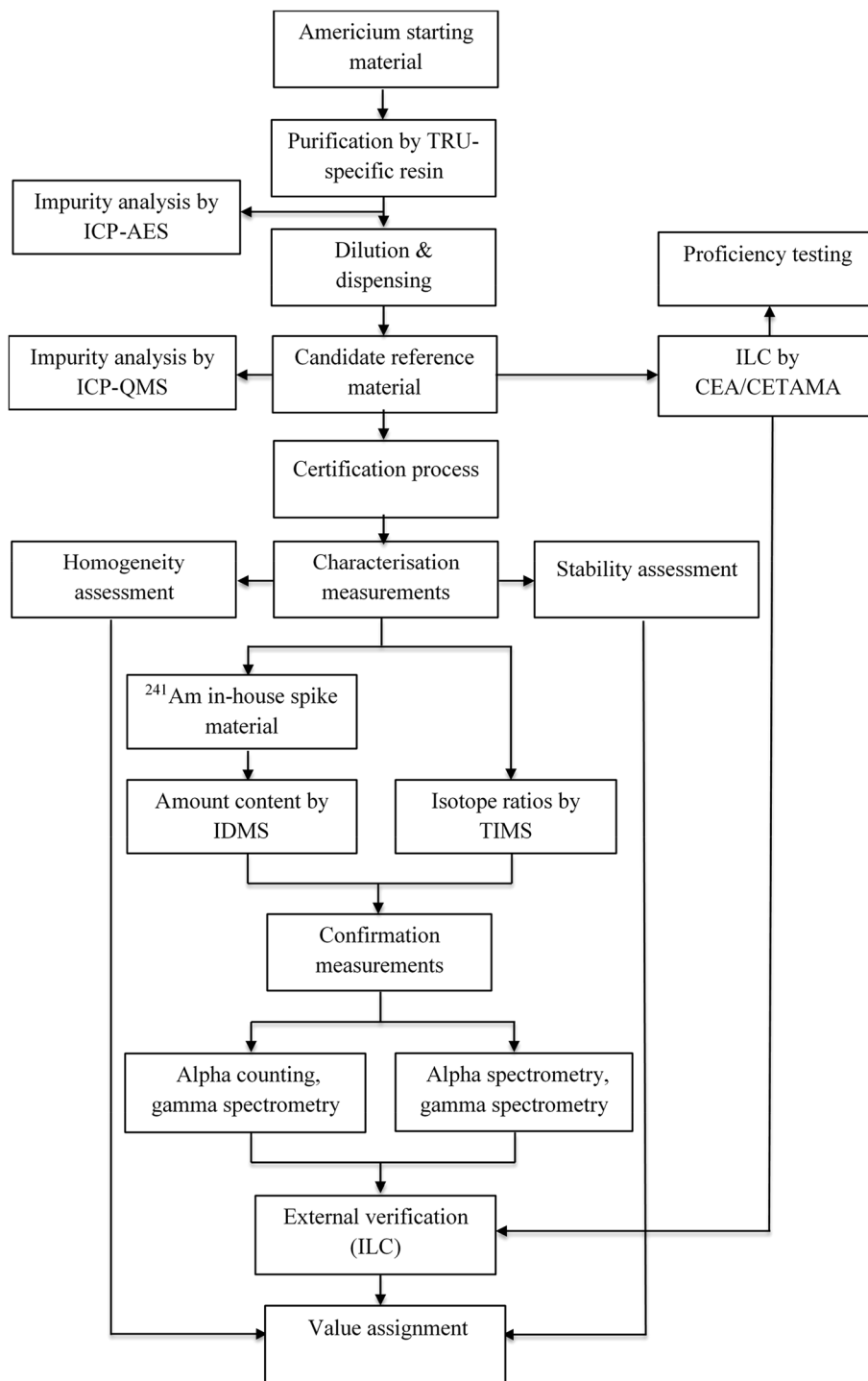
not available at JRC-Geel. On the other hand, the characterisation by alpha- or gamma spectrometry would result in higher uncertainty. The second strategy relies on the complete removal of the ^{241}Am in the ^{241}Pu material and setting the “clock” to zero (time zero). The amount of produced ^{241}Am by beta decay of ^{241}Pu (half-life 14.325 (24) years, $k = 2$ [18]) can be calculated at a given time using the appropriate Bateman equation [10]. The concentration of ^{241}Pu in the purified solution is determined by reverse IDMS using a certified plutonium spike CRM. This approach had previously been tested in a feasibility study [19]. For that study, a less enriched ^{241}Pu material (about 70%) originating from Oak Ridge National Laboratory was used for IDMS measurements of a similar ^{243}Am working reference material and was successfully applied in age determination of Pu NBL SRM 147 material. The lessons learned from the feasibility study were further developed and optimised for this certification project.

The $n(^{241}\text{Am})/n(^{243}\text{Am})$ and $n(^{242\text{m}}\text{Am})/n(^{243}\text{Am})$ amount ratios were measured by TIMS. Alpha-particle spectrometry, alpha-particle counting at a defined solid angle (DSA), and high-resolution gamma-ray spectrometry were applied as independent confirmation measurement techniques. Furthermore, the reference values obtained through the certification process were verified externally against the results of an interlaboratory comparison (ILC) organised by CEA/CETAMA, using this reference material as the test sample. The steps in the preparation and certification of the americium reference material are shown in Fig. 1.

Material production

Four milligrams of the Am starting material consisting of approximately 88% ^{243}Am , 12% ^{241}Am and 0.01% $^{242\text{m}}\text{Am}$ were purified by extraction chromatography to remove the major elemental impurities and subsequently diluted with about 2400 mL 1 M nitric acid solution to obtain an Am mass fraction of $1.5 \mu\text{g g}^{-1}$ solution. The solution was dispensed into pre-cleaned glass ampoules with screw-caps. In total 587 units of this reference material were prepared, each unit containing approximately 5 μg of Am in 3.5 mL nitric acid solution. Impurity analyses were carried out on the purified Am starting solution by inductively coupled plasma atomic emission spectroscopy (ICP-AES), as well as on the final product by inductively coupled plasma quadrupole mass spectrometry (ICP-QMS). All labware (flask, dispenser and ampoules) were cleaned prior to use with ultra-pure Milli Q water (resistivity 18.2 M Ω cm at 25 °C) and 10% (m/m) nitric acid solution for 24 h and dried in an oven at 60 °C [20].

Fig. 1 Flow chart for the preparation and certification of IRMM-0243/STAM



Material characterisation

$^{241}\text{Am}/^{241}\text{Pu}$ in-house spike

About 2 milligrams of ^{241}Pu material were dissolved in nitric acid and purified by anion exchange separation (Bio-rad AG 1-X4 resin, 100–200 mesh) [21] to remove the daughter decay products. The purification procedure was

performed three times in order to remove all the decay products that had been growing in since the production of this material in 1991. The final purification was carried out on June 10, 2014 at 15:10 CET (Central European Time) and marked the start of the ^{241}Am in-growth (time zero). An aliquot of the purified ^{241}Pu solution was measured by high-resolution gamma-ray spectrometry to verify the completeness of the purification; the atom ratio

$^{241}\text{Am}/^{241}\text{Pu}$ was found to be less than $5.6 (6) \times 10^{-5}$ at time zero.

The purified ^{241}Pu material was diluted with 100 mL 1 M nitric acid solution and characterised for the amount content of ^{241}Pu and for isotopic composition. The weight of the flask containing the ^{241}Pu solution was monitored to correct for any evaporation loss.

Spiking

Spiking, chemical purifications, and subsequent measurements were carried out about one and a half year after the purification of the ^{241}Pu material (June 10, 2014). During this period a sufficient amount of ^{241}Am had been produced (about 5.67 nmol g^{-1} at $t = 18$ months, see also Fig. 3) for spiking. Eighteen units in total were selected (stratified random) from the whole batch and from each unit an aliquot of about 2.5 g was mixed with an aliquot of about 1.0 g of the $^{241}\text{Pu}/^{241}\text{Am}$ spike solution (blend) for the determination of the amount content of ^{243}Am by IDMS. In addition an aliquot of about 0.7 g was taken from each ampoule for the determination of the $n(^{241}\text{Am})/n(^{243}\text{Am})$ and $n(^{242\text{m}}\text{Am})/n(^{243}\text{Am})$ isotope ratios by TIMS. Spiking was done by metrological weighing using the substitution method [22]. The chemical separation and isotope ratio measurements were carried on sets of two units over a period of 7 months (January to July 2016). This approach was used in order to have different amounts of ingrown ^{241}Am for spiking and to assess the stability of the Am solution during the certification campaign.

Americium purification

Prior to the isotope ratio measurements, two purification steps were carried out to remove the ^{241}Pu from the in-grown ^{241}Am in the blends by means of UTEVA-spec [23] and DGA extraction resins [24] (both of Triskem International, Bruz, France). The time of the Am elution (i.e. separation of Am from Pu) from the first UTEVA-spec column was carefully recorded in order to calculate the ingrowth time $\Delta t(^{241}\text{Am})$, i.e. the time elapsed since time zero. The DGA purification step was applied to remove the organic and other impurities that would interfere in the isotopic measurements. The purification steps of Am prior to isotopic measurement are shown in Fig. 2.

Isotope ratio measurements

Isotope ratio measurements were carried out using the total evaporation method on a multi-collector Triton Thermal Ionisation Mass Spectrometer (Thermo Fisher Scientific, Bremen, Germany), in a similar manner as routinely performed for uranium (U) and plutonium (Pu) samples of similar size. In the total evaporation method,

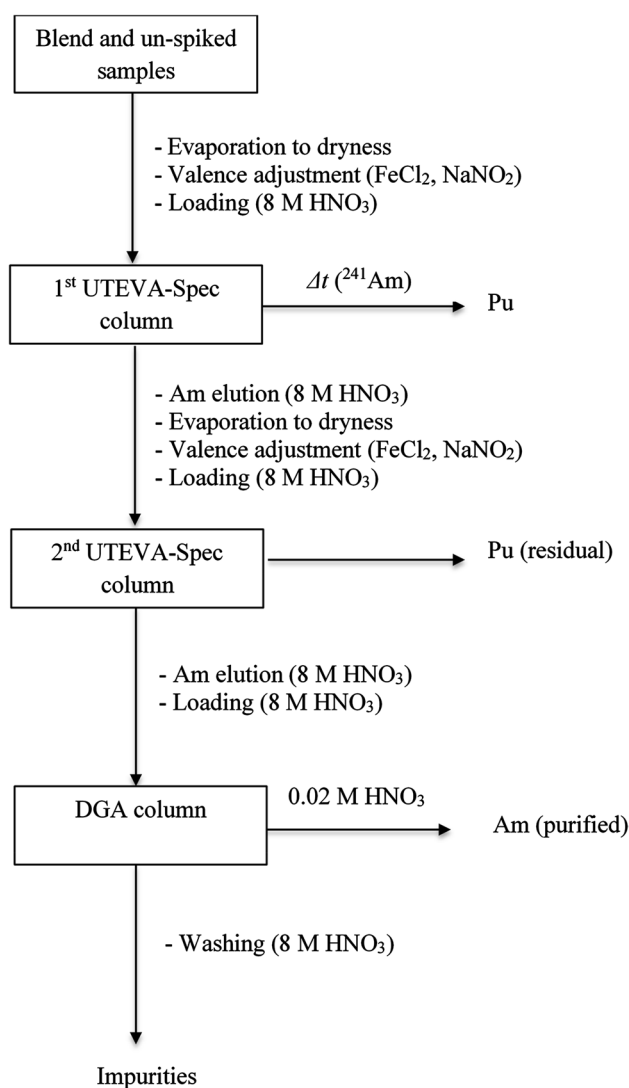


Fig. 2 Purification of Am prior to isotope ratio measurement

the evaporation filament is heated up to maintain a steady intensity and measured until the whole sample is consumed. In this way, the fractionation effects in the ion source are minimized [25–29]. A mass fractionation correction by measuring an americium isotopic standard was not possible due to unavailability of such material. It can be assumed that Am behaves similar to U and Pu during the total evaporation measurements (similar first ionisation potentials) [30]. Therefore, Pu isotopic standards were measured to check the performance of the mass spectrometer. Degassed zone refined rhenium filaments (Thermo Fisher Scientific, Bremen, Germany) were used as ionization and evaporation filaments (double filament configuration). About 20 ng of Am in the form of nitrate solution were deposited on an evaporation filament, dried down and mounted on a sample turret. All ion currents were measured simultaneously on Faraday cups.

Confirmation measurements

High-resolution alpha-particle spectrometry [31–33] was used to determine the activity ratio $A(^{243}\text{Am})/A(^{241}\text{Am})$ of the Am reference solution. The alpha source was prepared by electrodeposition on a polished stainless steel disk with an active diameter of 18.6 mm and measured using a passivated ion-implanted planar silicon detector (PIPS[®], 150 mm² active area, Mirion Technologies (MGPI) SA, France). The total Am activity per unit mass was determined by means of alpha-particle counting at a defined solid angle (DSA) [31, 34]. Gravimetrically quantified drops of Am solution were deposited on 34 mm glass plates and covered with 20 $\mu\text{g cm}^{-2}$ VYNS foils (polyvinylchloride-polyvinylacetate copolymer) to prevent material loss.

Two independent gamma-ray measurement campaigns were carried out, one in the underground laboratory ‘HADES’ [35] and one above ground in the radionuclide metrology laboratory at the JRC-Geel. Point-like sources were prepared gravimetrically by drop deposition on laminated plastic foils supported by a stainless steel annulus of 34 mm outer diameter. They were measured above ground using two coaxial high purity germanium (HPGe) gamma-ray spectrometers, one of 35% and the other of 90% relative efficiency [Mirion Technologies (MGPI) SA, France]. One gamma source was selected for ultra-low-level gamma-ray spectrometry (ULGS) measurements in the ‘HADES’ laboratory using a HPGe detector of type Broad Energy Germanium Detector (BEGe) with a relative efficiency of 20% (Canberra, USA). The activities of ²⁴¹Am and ²⁴³Am were calculated based on the main gamma-ray peaks of the nuclides, i.e. at 59.54 keV energy for ²⁴¹Am and at 74.66 keV for ²⁴³Am [36, 37].

Results and discussion

²⁴¹Am/²⁴¹Pu in-house spike

The results of the characterisation measurements of the ²⁴¹Pu solution after purification are summarised in Table 1.

The values for the isotope ratios were found to be in agreement with the values from the measurement certificate of the ²⁴¹Pu material of 1991. The ²⁴¹Pu content (i.e. mass fraction, amount content) was determined as the mean of the 10 blends measured by IDMS using ²⁴²Pu spike CRM (IRMM-049d). Each blend was measured in replicates on the Triton TIMS using the total evaporation method. The correction for mass fractionation effects was based on the measurement of the IRMM-290b/A3 Pu isotopic standard.

The ingrown amount of the ²⁴¹Am ($c_{241\text{Am}}$) was determined using the equation (Eq. 1):

Table 1 ²⁴¹Pu amount content, ²⁴¹Pu mass fraction, isotope amount ratios and associated uncertainties in the purified ²⁴¹Pu solution

Description (unit)	Measurand	Value
Amount content (mol g ⁻¹)	$c(^{241}\text{Pu})$	$8.1394 (53) \times 10^{-8}$
Mass fraction (g g ⁻¹)	$\gamma (^{241}\text{Pu})$	$1.9621 (13) \times 10^{-5}$
Amount ratios (mol mol ⁻¹)	$n(^{238}\text{Pu})/n(^{241}\text{Pu})$	$9.8 (34) \times 10^{-6}$
	$n(^{239}\text{Pu})/n(^{241}\text{Pu})$	$5.3 (19) \times 10^{-6}$
	$n(^{240}\text{Pu})/n(^{241}\text{Pu})$	$2.5923 (29) \times 10^{-3}$
	$n(^{242}\text{Pu})/n(^{241}\text{Pu})$	$4.4427 (25) \times 10^{-3}$

The reference date of the values is June 10, 2014

$$c_{241\text{Am}} = c_{241\text{Pu}}^o \frac{\lambda_{241\text{Pu}}}{\lambda_{241\text{Am}} - \lambda_{241\text{Pu}}} (e^{-\lambda_{241\text{Pu}} \Delta t} - e^{-\lambda_{241\text{Am}} \Delta t}) + c_{241\text{Am}}^o e^{-\lambda_{241\text{Am}} \Delta t} \quad (1)$$

where $c_{241\text{Pu}}^o$ is the amount content [mol g⁻¹] of ²⁴¹Pu in the purified spike solution at time zero (June 10, 2014), Δt [a] is the ingrowth time of ²⁴¹Am (time elapsed since time zero), $\lambda_{241\text{Pu}}$ and $\lambda_{241\text{Am}}$ are the decay constants [a⁻¹] for ²⁴¹Pu and ²⁴¹Am, respectively, and $c_{241\text{Am}}^o$ is the initial amount content [mol g⁻¹] of ²⁴¹Am in the purified spike solution. The half-lives and associated uncertainties used for the calculation of the respective decay constants were 14.325 (24) years for ²⁴¹Pu and 432.6 (1.2) years for ²⁴¹Am [1, 18].

To determine any ²⁴¹Am impurity left in the ²⁴¹Pu solution after purification, the ²⁴¹Am ingrowth in the ²⁴¹Pu solution was followed by high-resolution gamma-ray spectrometry over a longer period. This indicated a separation time of June 10, 2014 17:18 CET (uncertainty range from 10:10 to 23:55). This agrees very well with the actual separation time of June 10, 2014, 15:10 CET. Based on these measurements, the initial amount of ²⁴¹Am in the purified ²⁴¹Pu solution was considered to be negligible and therefore $c_{241\text{Am}}^o \approx 0$.

The ingrowth of ²⁴¹Am in the ²⁴¹Pu solution after the purification is shown as a mass fraction in Fig. 3.

Impurities in the final reference material

The impurities measured in the reference material solution by ICP-QMS using a Perkin Elmer Elan instrument were potassium (Na), lead (Pb), uranium (²³⁸U) and zinc (Zn) with mass fractions of 5.3 (2.7) $\mu\text{g g}^{-1}$, 0.96 (53) $\mu\text{g g}^{-1}$, 1.62 (81) $\mu\text{g g}^{-1}$ and 1.48 (75) $\mu\text{g g}^{-1}$, respectively.

Furthermore, ²⁴²Cm and ²⁴³⁺²⁴⁴Cm radioisotopes were measured by alpha spectrometry with activity of 59 (6) Bq g⁻¹ and 12 (1) Bq g⁻¹, respectively. The contribution of ²⁴²Cm isobaric impurity ($5 \times 10^{-7} \mu\text{g g}^{-1}$) to the fraction of ^{242m}Am in the reference material was about 0.2%. It was included in the uncertainty budget of the $n(^{242\text{m}}\text{Am})/n(^{243}\text{Am})$ ratio.

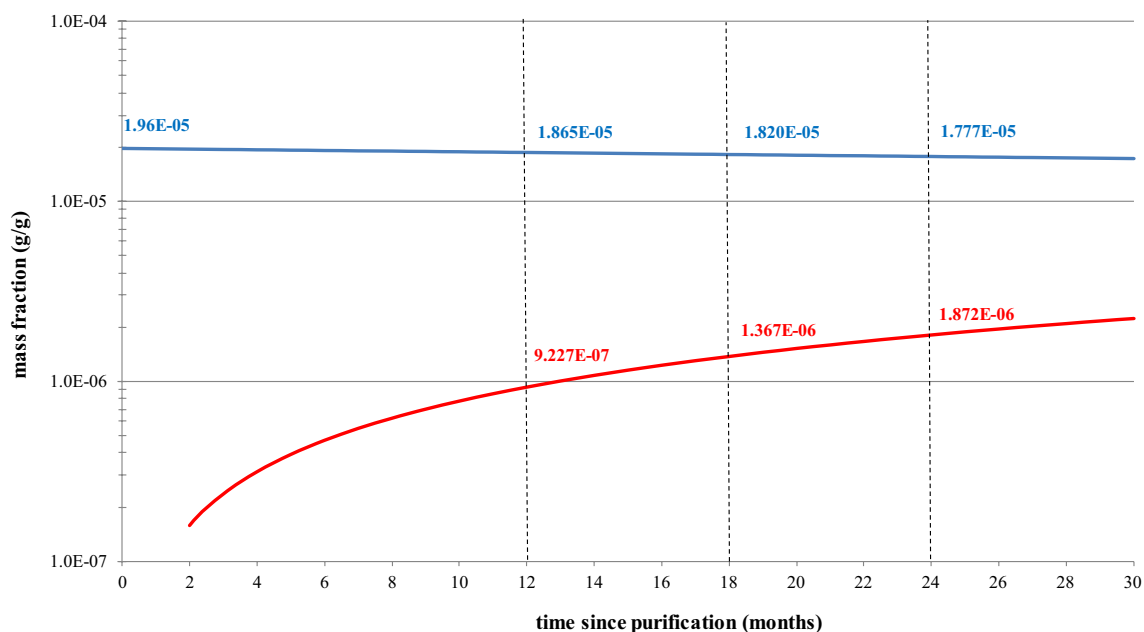


Fig. 3 Ingrowth of ^{241}Am (red curve) in the ^{241}Pu solution (blue line) after the purification (time zero is June 10, 2014). (Color figure online)

Assigned values and uncertainty estimation

The amount content of ^{243}Am in the reference material was determined as the mean of sixteen results obtained by IDMS on the selected units. Two units (unit no. 240 and 437) were excluded from the evaluation due to very low signal intensities during the isotope measurement.

The $n(^{241}\text{Am})/n(^{243}\text{Am})$ and $n(^{242\text{m}}\text{Am})/n(^{243}\text{Am})$ ratios were measured on a Triton TIMS using the total evaporation method. The results for the amount content of ^{243}Am and the $n(^{241}\text{Am})/n(^{243}\text{Am})$ ratio are shown in Figs. 4 and 5, respectively. The amount contents of the ^{241}Am , the total Am and the corresponding mass fractions were derived from the ^{243}Am amount content, the $n(^{241}\text{Am})/n(^{243}\text{Am})$

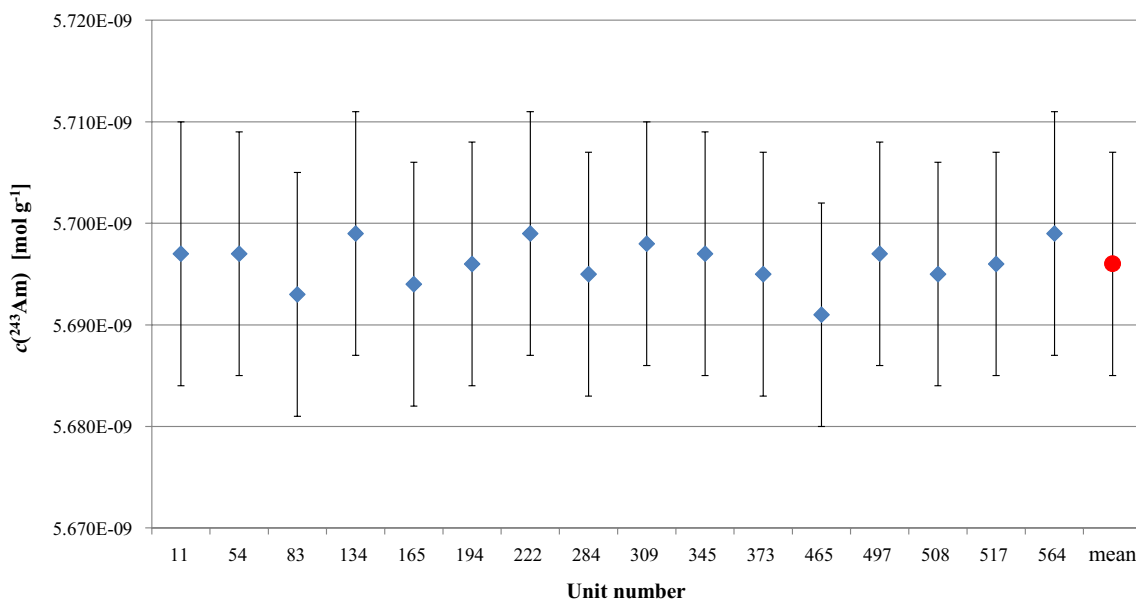


Fig. 4 The amount content of ^{243}Am in the selected units of IRMM-0243 (blue diamonds) and the mean (red circle) determined by IDMS. Error bars represent the expanded ($k=2$) uncertainties. (Color figure online)

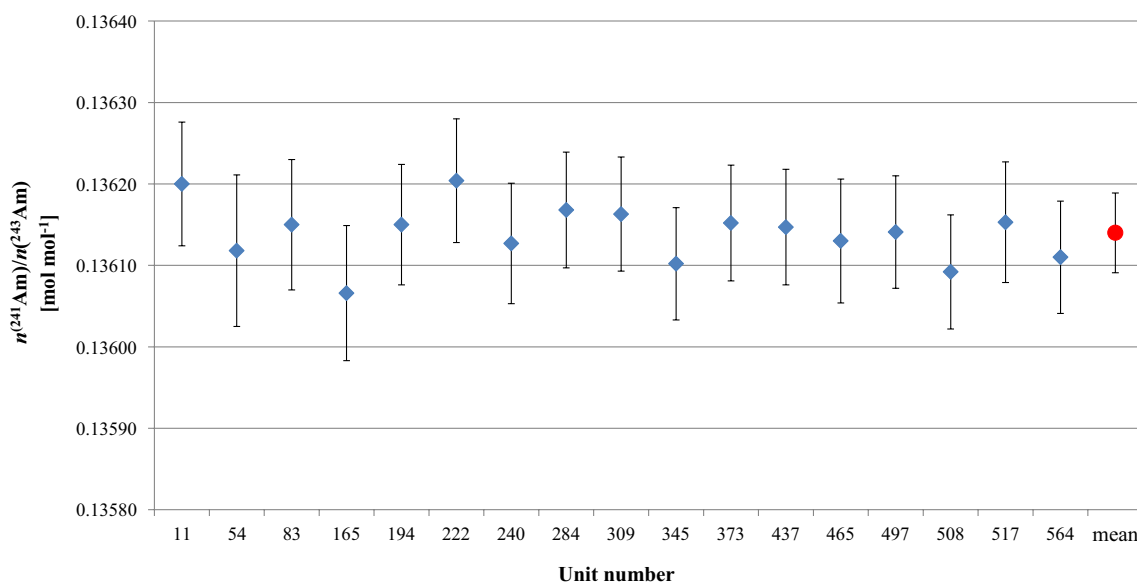


Fig. 5 The $n(^{241}\text{Am})/n(^{243}\text{Am})$ ratio in the selected units of IRMM-0243 (blue diamonds) and the mean (red circle) determined by TIMS. Error bars represent the expanded ($k=2$) uncertainties. (Color figure online)

amount ratio, and the respective atomic masses [38]. Measurement uncertainties were evaluated according to the Guide to the Expression of Uncertainty in Measurement (GUM) [39] using the GUM Workbench© software [40].

A mass fractionation correction using measurements of a CRM on the same magazine as the sample is recommended following ASTM C1672-17, but not mandatory [28]. This correction was not possible to perform for the Am measurements for this project, since there was no suitable Am isotopic standard available. Due to similarities in the chemical behaviour and similar ionization energies, it can be assumed that Am behaves similar to U or Pu during the total evaporation measurement and that the bias statements for U and Pu can equally be applied for americium. For the $n(^{241}\text{Am})/n(^{243}\text{Am})$ ratio the uncertainty component was calculated as 0.033% ($k=2$, for ratios spanning 2 mass units) and for the $n(^{242\text{m}}\text{Am})/n(^{243}\text{Am})$ ratio to 0.017% ($k=2$, for ratios spanning 1 mass unit). These two uncertainty components were included in the combined uncertainty for isotope measurements. In addition, an uncertainty of 0.020% ($k=2$, for ratios spanning 2 mass units) derived from isotope measurements of the U and Pu quality control (QC) samples was added as a conservative contribution to the uncertainty budget for Am. In the case of $n(^{242\text{m}}\text{Am})/n(^{243}\text{Am})$ amount ratio, an additional source of uncertainty (0.2% rel.) was added to account for the ^{242}Cm impurity in the IRMM-0243 material. Relative contributions of various uncertainty components for the amount content of ^{243}Am and the $n(^{241}\text{Am})/n(^{243}\text{Am})$ ratio are shown in Figs. 6 and 7, respectively.

The relative expanded uncertainty (coverage factor $k=2$) for the ^{243}Am amount content from the assessment was 0.19%. The dominant source of uncertainty comes from the uncertainty of the half-life of ^{241}Pu (rel. contribution of 73.5%) followed by the concentration of the ^{241}Pu spike solution (rel. contribution of 12.3%). Other uncertainty components come from the ^{241}Am ingrowth time, the between-unit variability (homogeneity assessment) and isotope ratio measurements.

The uncertainty of the $n(^{241}\text{Am})/n(^{243}\text{Am})$ ratio consists of the repeatability of the isotope measurements and uncertainty components from the potential bias during the isotope ratio measurement and the QC measurements.

The assigned values and the results from the confirmation measurements (i.e. alpha and gamma spectrometry) and the results from the ILC contributing to the certification of STAM/IRMM-0243 are summarised in Table 2. An agreement was achieved within measurement uncertainties for all the measurands in Table 2 [39, 41]. In particular, good agreement was obtained for the results from alpha spectrometry and the reported results (as weighted mean) in the ILC. The compatibility between the assigned reference values and the ILC was quantified by the normalised deviation (E_n score) according to ISO 13528 [42]. The normalised deviations for the amounts of ^{243}Am and ^{241}Am and the $n(^{241}\text{Am})/n(^{243}\text{Am})$ ratio, -0.3 , 0.1 and 0.8 , respectively, were all less than 2 [15]. Therefore, the values were adopted as the certified values for IRMM-0243/STAM.

In the case of the $n(^{242\text{m}}\text{Am})/n(^{243}\text{Am})$ ratio, a value of 0.0001506 (15) was measured by TIMS. This value was higher than the value obtained by the ILC, 0.0001367 (9).

Fig. 6 Relative uncertainty contributions (%) from the characterisation of the amount content of ^{243}Am (top pie chart). The bottom pie chart shows the relative uncertainty contributions of the ^{241}Pu solution

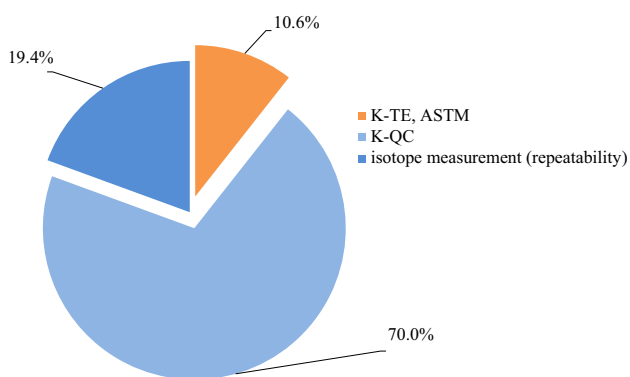
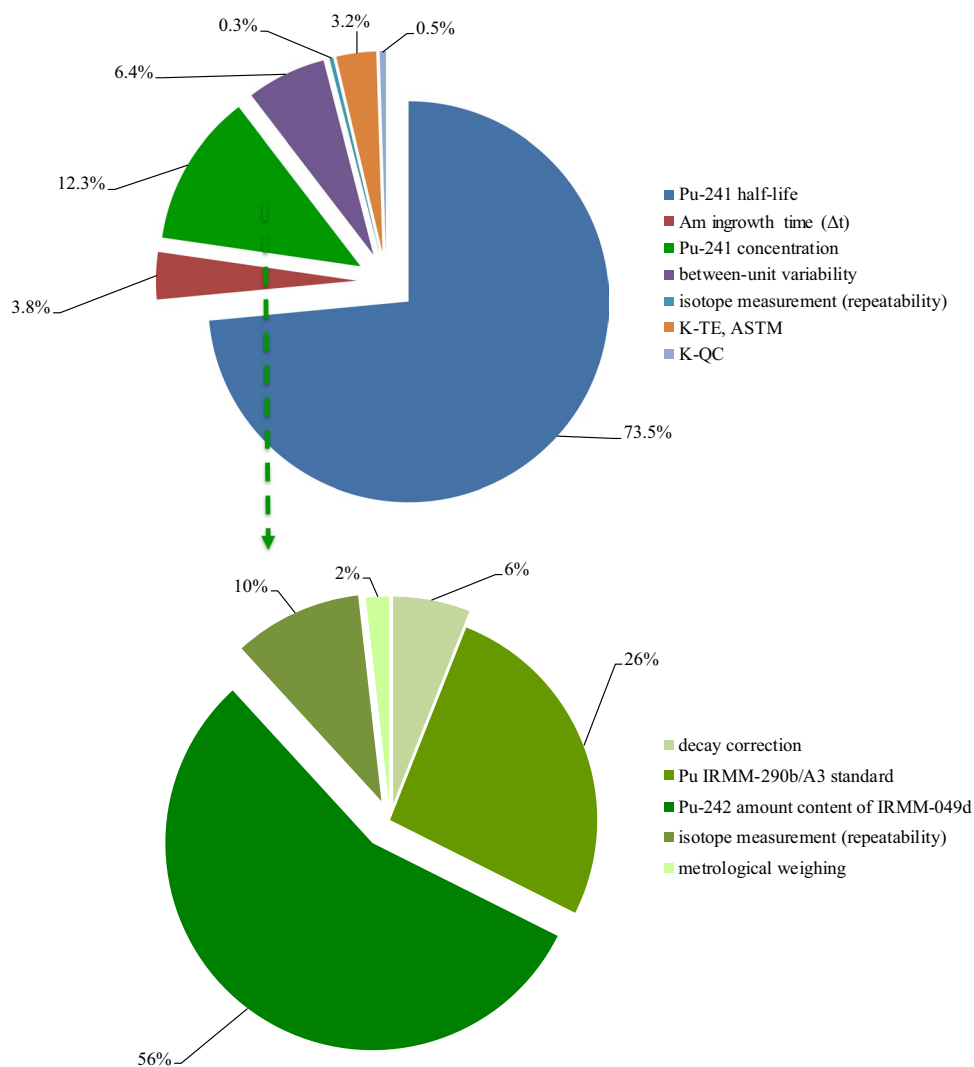


Fig. 7 Relative uncertainty contributions (%) from the characterisation of the $n(^{241}\text{Am})/n(^{243}\text{Am})$ ratio

Additional measurements showed several hundreds of counts at masses 238, 239 and 242, which may explain this observation. These interferences came from the reagents, resins or

other impurities that were not removed by chemical purification prior to the measurement. The $n(^{242\text{m}}\text{Am})/n(^{243}\text{Am})$ ratio could not be corrected quantitatively for this effect and, therefore only an indicative value based on the ILC results has been assigned for this ratio.

Conclusions

A novel ^{243}Am spike reference material IRMM-0243 (called STAM at CEA/CETAMA) was prepared and certified for the amount content of ^{243}Am and the $n(^{241}\text{Am})/n(^{243}\text{Am})$ isotope amount ratio by IDMS and TIMS and successfully confirmed by independent radioactivity measurements. Indicative values for the $n(^{242\text{m}}\text{Am})/n(^{243}\text{Am})$ isotope amount ratio and its uncertainty were derived from the ILC organised by CEA/CETAMA. The material is supplied in a glass ampoule with a screw-cap containing about 3.5 mL of dilute nitric acid solution with an Am mass fraction of $1.5716(30) \mu\text{g g}^{-1}$.

Table 2 The assigned values, the results of the confirmation measurements and their uncertainties of the IRMM-243/STAM reference material

Measurand (unit)	Assigned value ^a	Confirmation measurements			
		Alpha spectrometry	Gamma spectrometry ^b	Gamma spectrometry ^c	ILC ^d
²⁴¹ Am (nmol g ⁻¹)	0.7754 (15)	0.7760 (33)	0.776 (9)	0.769 (17)	0.7756 (22)
²⁴³ Am (nmol g ⁻¹)	5.696 (11)	5.714 (40)	5.65 (12)	5.63 (24)	5.693 (16)
$n(^{241}\text{Am})/n(^{243}\text{Am})$	0.136138 (54)	0.1358 (11)	0.1375 (27)	0.137 (7)	0.13618 (8)
$n(^{242\text{m}}\text{Am})/n(^{243}\text{Am})$	/	/	/	/	0.0001367 (9)

The reference date of the values is January 1, 2017

^aCertified values

^bJRC-Geel radionuclide laboratory

^cHADES (underground laboratory)

^dWeighted mean [15]

This concentration is suitable for various mass spectrometry measurements using, e.g., TIMS, ICP-MS [43].

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References

- DDEP (2004–2020) DDEP Monographie BIPM-5—Table of Radionuclides <http://www.inhb.fr/nuclear-data/nuclear-data-table/> Accessed 4 Aug 2020
- Marouli M, Pommé S, Jobbagy V, Stroth H, Van Ammel R, Fankhauser A, Jakopic R, Richter S, Aregbe Y, Crozet M, Maillard C, Rivier C, Roudil D (2020) Absolute and relative measurement of the ²⁴³Am half-life. *J Radioanal Nucl Chem*. <https://doi.org/10.1007/S10967-020-07450-9>
- Vajda N, Kim CK (2010) Determination of ²⁴¹Am isotope: a review of analytical methodology. *J Radioanal Nucl Chem* 284:341–366
- Agarwal SK (2018) A review on the mass spectrometric studies of americium: present and future perspectives. *Mass Spectrom Rev* 37:43–56
- Balsley SD (2014) Report of the 2014 technical meeting on nuclear reference materials for destructive analysis in the nuclear fuel cycle. IAEA SAL, 2–4 September, Vienna
- The Nuclear Security Summit 2016. <http://www.nss2016.org/>
- Wallenius M, Peerani P, Koch L (2000) Origin determination of plutonium material in nuclear forensics. *J Radioanal Nucl Chem* 246:317–321
- Nygren U, Rameback H, Nilsson C (2007) Age determination of plutonium using inductively coupled plasma mass spectrometry. *J Radioanal Nucl Chem* 272:345–510
- Wallenius M, Mayer K (2000) Age determination of plutonium material in nuclear forensics by thermal ionisation mass spectrometry. *Fresenius J Anal Chem* 366:234–238
- Keegan RP, Gehrke RJ (2003) A method to determine the time since last purification of weapons grade plutonium. *Appl Radiat Isot* 59:137–143
- Byerly BL, Stanley F, Spencer K, Colletti L, Garduno K, Kuhn K, Lujan E, Martinez A, Portfield D, Rim J, Schappert M, Thomas M, Townsend L, Xu N, Tandon L (2016) Forensic investigation of plutonium metal: a case study of CRM 126. *J Radioanal Nucl Chem* 310:623–632
- Chartier F, Aubert M, Pilier M (1999) Determination of Am and Cm in spent nuclear fuels by isotope dilution inductively coupled plasma mass spectrometry and isotope dilution thermal ionization mass spectrometry after separation by high-performance liquid chromatography. *Fresenius J Anal Chem* 364:320–327
- ISO 17034:2016 (2016) General requirements for the competence of reference material producers. International Organization for Standardisation, Geneva
- ISO Guide 35 (2017) Reference materials—general and statistical principles for certification. International Organization for Standardization, Geneva
- Crozet M, Roudil D, Maillard C, Rivier C, Jakopic R, Fankhauser A, Aregbe Y, Richter S (2019) Contribution of an interlaboratory comparison to the certification of the STAM/IRMM-0243 ²⁴³Am reference material. *J Radioanal Nucl Chem* 319:717–725
- Vogl J, Pritzkow W (2010) Isotope dilution mass spectrometry—a primary method of measurement and its role for RM certification. *J Metrol Soc India* 25:135–164
- Milton MJ, Quinn TJ (2001) Primary methods for the measurement of amount of substance. *Metrologia* 38:289–296

18. Wellum R, Verbruggen A, Kessel R (2009) A new evaluation of the half-life of ^{241}Pu . *J Anal At Spectrom* 24:801–807
19. Sturm M (2017) Reference materials for age dating in nuclear forensics. Ph.D. dissertation, University of Natural Resources and Life Sciences (BOKU), Vienna
20. Jakopič R, Fanhauser A, Aregebe Y, Crozet M, Maillard C, Richter S, Rivier C, Roudil D, Altitzoglou T, Pommé S, Marouli M, Tzika F (2017) Preparation and certification of ^{243}Am spike reference material: IRMM-0243, certification report EUR 28748 EN
21. Jakopič R, Verbruggen A, Eykens R, Kehoe F, Kuhn H, Kushigeta J, Jacobsson U, Bauwens J, Richter S, Wellum R, Aregebe Y (2010) An inter-calibration campaign using various selected Pu spike isotopic reference materials. *J Radioanal Nucl Chem* 286:449–454
22. OIML R111-1 (2004) Weights of classes E1, E2, F1, F2, M1, M1-2, M2, M2-3 and M3, Part 1: Metrological and technical requirements. International Organisation of Legal Metrology, Paris
23. Morgenstern A, Apostolidis C, Carlos-Marquez R, Mayer K, Molinet R (2002) Single-column extraction chromatographic separation of U, Pu, Np and Am. *Radiochim Acta* 90:81–85
24. Kazi ZH, Cornett JR, Zhao X, Kieser L (2014) Americium and plutonium separation by extraction chromatography for determination by accelerator mass spectrometry. *Anal Chim Acta* 829:75–80
25. Callis EL, Abernathy RM (1991) High precision isotopic analysis of uranium and plutonium by total sample volatilization and signal integration. *Int J Mass Spectrom Ion Process* 103:322–327
26. Bürger S, Balsley SD, Baumann SD, Boulyga SF, Cunningham JA, Kappel S, Kopft A, Poths J (2012) Uranium and plutonium analysis of nuclear material samples by multi-collector thermal ionisation mass spectrometry: quality control, measurement uncertainty, and metrological traceability. *Int J Mass Spectrom* 311:40–50
27. Boulyga S, Konnegger-Kappel S, Richter S, Sangley L (2015) Mass spectrometry analysis for nuclear safeguards. *J Anal At Spectrom* 30:1469–1489
28. 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, ASTM International
29. Quemet A, Ruas A, Dalier V, Rivier C (2018) Americium isotope analysis by thermal ionisation mass spectrometry using the total evaporation method. *Int J Mass Spectrom* 431:8–14
30. Lide DR (1992) Ionization potentials of atoms and atomic ions. In: *Handb. Chem Phys* 10–211
31. Pommé S, Sibbens G (2008) Alpha-particle counting and Spectrometry in a primary Standardisation laboratory. *Acta Chim Slov* 52:111–119
32. Pommé S, Marroyo BC (2015) Improved peak shape fitting in alpha spectra. *Appl Radiat Isotopes* 96:148–153
33. Pommé S (2015) Typical uncertainties in alpha-particle spectrometry. *Metrologia* 52:S146–S155
34. Pommé S (2015) The uncertainty of counting at a defined solid angle. *Metrologia* 52:S73–S85
35. Hult M, Preusse W, Gasparro J, Kohler M (2006) Underground gamma-ray Spectrometry. *Acta Chim Slov* 53:1–7
36. Hult M (2007) Low-level gamma-ray spectrometry using Ge-detectors. *Metrologia* 44/4, S87–S94, Erratum at *Metrologia* 44/5, 425
37. Kawrakow I, Rogers DWO (2003) PIRS-701: the EGSnrc code system: Monte Carlo simulation of Electron and Photon Transport: Ionizing Radiation Standards. NRC, Ottawa, Ontario
38. Wang M, Audi G, Wapstra AH, Kondev FG, MacCormick M, Xu X, Pfeiffer B (2012) The AME2012 atomic mass evaluation (II). *Tables Gr Ref Chin Phys C* 36:1603–2014
39. ISO, IEC 98–3 (2016) Guide to the expression of uncertainty in Measurement (GUM 1995). International Organisation for Standardisation, Geneva
40. MetroData GmbH, GUM Workbench Version 2.4: Software Package for the Evaluation of Uncertainty. <http://www.metrodata.de>
41. Pommé S, Keightley J (2015) Determination of a reference value and its uncertainty through a power-moderated mean. *Metrologia* 52:S200–S212
42. ISO13528 (2015) Statistical methods for use in proficiency testing by interlaboratory comparison. International Organisation for Standardisation, Geneva
43. <https://ec.europa.eu/jrc/en/reference-materials/catalogue>

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