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²⁴³Am certified reference material for nuclear safeguards and security

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Abstract:

The Joint Research Centre (EC-JRC, Geel, Belgium) and the Commissariat à l'Energie Atomique et aux Energies Alternatives (CEA/DEN, Marcoule, France) jointly prepared and certified a ²⁴³Am spike reference material in compliance with ISO 17034. This reference material is needed for accurate mass spectrometry measurements of ²⁴¹Am in nuclear forensics, security and safeguards applications but also for the characterisation of radioactive waste. Nearly 600 units of this material were produced, each containing about 3.5 mL of dilute nitric acid solution with an Am mass fraction of 1.5 µg·g⁻¹. The reference material is certified for the ²⁴³Am amount content by Isotope Dilution Mass Spectrometry (IDMS) using an in-house ²⁴¹Am spike, produced by beta-decay from highly enriched ²⁴¹Pu (99.3 %) material. The americium isotope amount ratios were certified by Thermal Ionisation Mass spectrometry (TIMS). Independent verification measurements were performed by alpha-particle spectrometry, high-resolution gamma-ray spectrometry for activity ratios, and alpha counting for the total massic activity of ^{241,243}Am.

Keywords: Americium; Certified Reference Material; Nuclear Safeguards; Nuclear Forensics; IDMS; TIMS; ISO 17034; ISO Guide 35

1. Introduction

In nuclear safeguards and security, accurate isotopic measurements are required in order to draw correct conclusions. The accuracy, reliability and traceability of such measurements depend heavily on suitable isotopic reference materials. There is a wide range of uranium and plutonium Certified Reference Materials (CRMs) for quality control, method validation and instrument calibration in mass spectrometry. On the contrary, the availability of americium reference materials is limited. Currently, there is no ²⁴³Am spike reference material commercially available, although a certified reference material is indispensable for accurate mass spectrometry measurements of ²⁴¹Am in nuclear materials. Such material can be used in nuclear forensics to determine the 'model age' of a (seized) plutonium material, i.e. the time elapsed since its last chemical purification [1, 2, 3, 4]. Accurate measurements of elemental americium and isotopic composition are also needed for the management of nuclear waste, where ²⁴¹Am contributes via its daughter ²³⁷Np to the long-lived radioactive waste [5].

The provision of nuclear reference materials is regularly addressed among reference materials providers and users, e.g. in the frame of the Working Group on Techniques and Standards for Destructive Analysis (WGDA) of the European Safeguards and Research Association (ESARDA), the International Atomic Energy Agency (IAEA), and the Nuclear Forensics International Technical

Working Group (ITWG). The need for an americium spike CRM was expressed at the 2014 IAEA Technical Meeting on Reference Materials for Destructive Analysis in the Nuclear Fuel Cycle and at the 2016 Nuclear Security Summit: Certified Reference Material Fact Sheet [6]. In order to fulfil this urgent need, a novel ^{243}Am spike material was jointly produced and certified by the Joint Research Centre of the European Commission (EC-JRC) in Geel (Belgium) and the Commissariat à l'Energie Atomique et aux Energies Alternatives (CEA/DEN) in Marcoule (France). The reference material was produced in compliance with ISO 17034 [7] and characterised for the ^{243}Am , ^{241}Am and total Am amount contents by Isotope Dilution Mass Spectrometry (IDMS) [8] and for the $n(^{241}\text{Am})/n(^{243}\text{Am})$ and $n(^{242\text{m}}\text{Am})/n(^{243}\text{Am})$ amount ratios by Thermal Ionisation Mass Spectrometry (TIMS). Furthermore, confirmation measurements of the certified values were performed by independent alpha-particle and gamma-ray spectrometry, as well as alpha-particle counting at a defined solid angle (DSA). Prior to release of this CRM, an inter-laboratory comparison (ILC) exercise using that same material has been organised by CEA/CETAMA.

The preparation and the characterisation of this ^{243}Am spike CRM will be presented in this paper. As the ILC exercise is still on-going, the certified values cannot be disclosed at this time. Therefore only the normalised results will be presented.

2. Preparation of the ^{243}Am reference material

Four mg americium source material (88 % ^{243}Am and 12 % ^{241}Am) was made available by the CEA/L2AT (ATalante Analysis Laboratory). The solution was purified by TRU-Spec resin (Triskem, France) to remove the impurities and shipped to JRC-Geel for processing. The purified americium solution was diluted with 2400 mL nitric acid solution ($[\text{HNO}_3] = 1 \text{ M}$) to achieve an Am mass fraction of $1.5 \mu\text{g}\cdot\text{g}^{-1}$. This Am concentration was considered suitable for various mass spectrometry measurements (e.g. TIMS, ICP-MS). The solution was allowed enough time to homogenise before being dispensed into pre-clean screw-cap ampoules. In total, 587 units were prepared, each unit containing approximately $5 \mu\text{g}$ of americium in nitric acid solution. Dilution of the original material and dispensing into ampoules were carried out in a dedicated glove box. The ampoules were packed in PVC bags and labelled. The major processing steps are depicted in Figure 1.

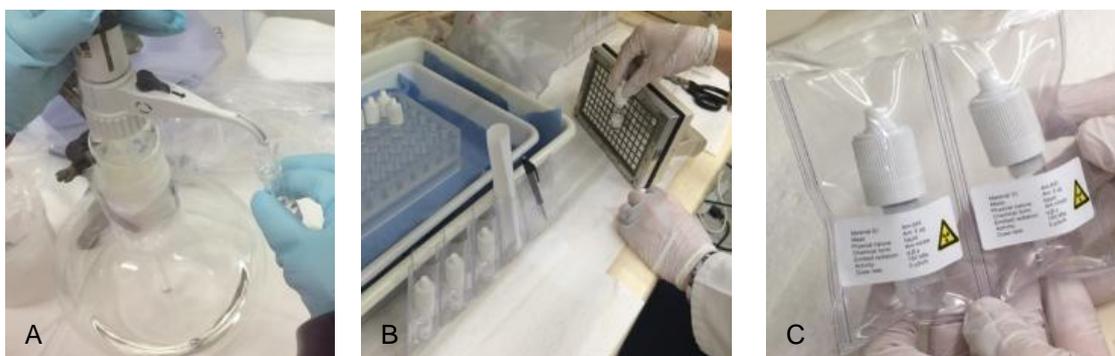


Figure 1: Preparation of ^{243}Am reference material: dilution and dispensing (A), packing and labelling (B and C)

3. Certification methodology

The ^{243}Am Am amount content was measured by Isotope Dilution Mass Spectrometry (IDMS). In IDMS, the amount of an element in the sample is determined on the basis of additions of known amounts of spike material of the same element, but with a significantly different isotopic composition from that of the unknown sample. By measuring the change in isotopic composition of the blend (sample-spike mixture), the unknown amount of the element in the sample can be determined [8].

The amount content of ^{243}Am ($C_{243\text{Am}}$) can be calculated using the following equation:

$$c_{243Am} = \frac{R_y - R_b}{R_b - R_x} \cdot R_x \cdot \frac{m_y}{m_x} \cdot c_{241Am}$$

Equation 1

where c_{241Am} is the ^{241}Am amount content ($\text{mol}\cdot\text{g}^{-1}$) of the spike; m_x and m_y are the mass (g) of the sample and the spike, respectively; R_i is the isotope amount ratio $n(^{243}\text{Am})/n(^{241}\text{Am})$ in which index i takes the values x , y and b to represent the un-spiked sample, the spike and the blend, respectively.

The $n(^{241}\text{Am})/n(^{243}\text{Am})$ and $n(^{242m}\text{Am})/n(^{243}\text{Am})$ amount ratios were determined by Thermal Ionisation Mass Spectrometry (TIMS). The material was certified in compliance with ISO 17034 [7] and ISO Guide 35 [9]. Homogeneity assessment and characterisation (value assignment) measurements were combined and carried out on 18 randomly selected units representing the whole batch.

3.1 ^{241}Am in-house spike

In absence of a suitable ^{241}Am spike material for IDMS measurements of the ^{243}Am , an alternative approach was applied. The ^{241}Am spike material was produced from highly enriched ^{241}Pu (99.3 %) material available at JRC-Geel. The ingrown ^{241}Am , produced by the beta-decay of ^{241}Pu since the initial purification of the ^{241}Pu material, was used as spike for the measurement of the ^{243}Am by IDMS.

About 2 mg of ^{241}Pu material was purified to remove the daughter decay products growing in since the production of this material in 1991. This was accomplished by anion exchange separation (Biorad AG 1-X4 resin, 100-200 mesh) [10]. The purification procedure was performed three times; the final purification was carried out on 10 June 2014 at 15:10 CET (Central European Time), which marked the start of the Am in-growth (time zero). The completeness of the purification was confirmed by high resolution gamma-ray spectrometry. The purified plutonium material was diluted with 100 mL nitric acid solution ($[\text{HNO}_3] = 1 \text{ M}$) and characterised for the ^{241}Pu amount content and its isotopic composition by IDMS using a ^{242}Pu spike CRM (IRMM-049d) and by Triton TIMS, respectively.

The ingrown amount of the ^{241}Am in ^{241}Pu is calculated using the following equation:

$$c_{241Am} = c_{241Pu}^0 \left[\frac{\lambda_{241Pu}}{\lambda_{241Am} - \lambda_{241Pu}} (e^{-\lambda_{241Pu} t} - e^{-\lambda_{241Am} t}) \right]$$

Equation 2

where c_{241Pu}^0 is the amount content ($\text{mol}\cdot\text{g}^{-1}$) of ^{241}Pu in the spike solution at time zero, t (a) is the ingrowth time of ^{241}Am (time elapsed since time zero) and λ_{241Am} and λ_{241Pu} are the decay constants (a^{-1}) for ^{241}Am and ^{241}Pu , respectively. The half-lives and associated uncertainties used in the calculations were $(14.325 \pm 0.012) \text{ a}$ ($k = 1$) [11] for ^{241}Pu and $(432.6 \pm 0.6) \text{ a}$ ($k = 1$) [12] for ^{241}Am .

3.2 Spiking and chemical treatment

Spiking, chemical treatment and subsequent measurements were carried out about one and a half year after the purification of the ^{241}Pu material. During this period a sufficient amount of ^{241}Am had been produced for IDMS analysis. From each of the 18 selected units, an aliquot was taken for determination of the ^{243}Am content by IDMS and another aliquot for the determination of the isotopic composition by TIMS. The chemical treatment and subsequent measurements were spread over a period of 7 months. This approach was used in order to have different amounts of ingrown ^{241}Am in the blends for IDMS and to be able to assess the stability of the ^{243}Am solution during the certification campaign. Prior to the measurements, two purification steps were carried out to remove the ^{241}Pu from the in-grown ^{241}Am in the spike solution by means of UTEVA-spec (Triskem, France) and DGA extraction resins (Triskem, France), respectively [13, 14]. The chemical procedure is shown in Figure 2.

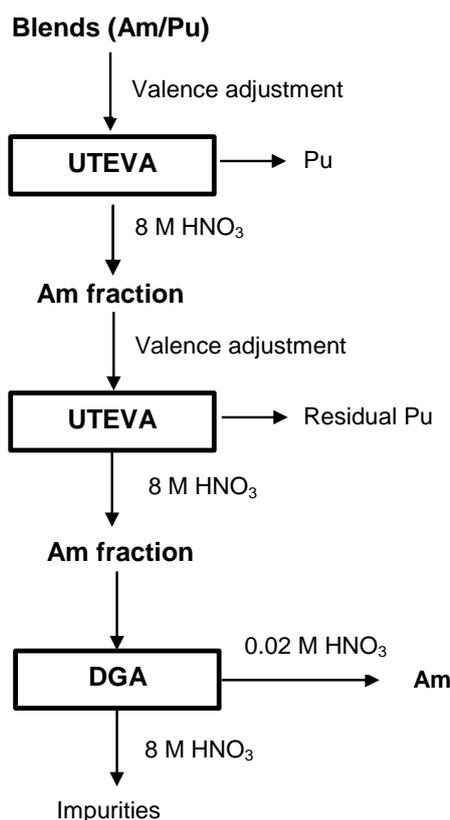


Figure 2: Purification of Am prior to isotope ratio measurement.

3.3 Isotope ratio measurement

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 and plutonium samples of similar size. In the total evaporation method, 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 [15, 16, 17, 18, 19]. Degassed zone refined rhenium filaments (Thermo Fisher Scientific, Bremen, Germany) were used as ionization and evaporation filaments (double filament configuration). About 20 ng of americium as nitrate solution was deposited on an evaporation filament, dried down and mounted on a sample turret. All ion currents were measured simultaneously on Faraday cups.

For U and Pu a mass fractionation correction using measurements of a CRM on the same magazine is recommended following ASTM C1672-17 [20], but not mandatory. According to ASTM C1672-17, the relative bias for uncorrected $n(^{235}\text{U})/n(^{238}\text{U})$ and $n(^{242}\text{Pu})/n(^{239}\text{Pu})$ ratios is less than 0.05 % within 2 standard deviations (2σ) for ratios spanning 3 mass units. In the absence of a suitable Am isotopic standard, it was not possible to perform a mass fractionation correction for this project. Due to similarities in the chemical behaviour and similar ionization energies, it can be assumed that americium behaves similar to uranium or plutonium during the total evaporation measurement and that the bias statements for uranium and plutonium can equally be applied for americium. For the $n(^{243}\text{Am})/n(^{241}\text{Am})$ ratio the uncertainty component was calculated as 0.033 % (2σ , for ratios spanning 2 mass units) and for the $n(^{242\text{m}}\text{Am})/n(^{243}\text{Am})$ ratio to 0.017 % (2σ , for ratios spanning 1 mass unit).

3.4 Verification measurements

Independent verification measurements for the ^{243}Am and ^{241}Am amount contents and for the $n(^{241}\text{Am})/n(^{243}\text{Am})$ amount ratios were performed using radioactivity measurement methods.

High-resolution alpha particle spectrometry [21, 22, 23] was used to determine the $A(^{243}\text{Am})/A(^{241}\text{Am})$ activity ratio. The alpha source was prepared from the Am CRM solution by electrodeposition on a polished stainless 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) [21, 24]. Gravimetrically quantified drops of americium solution were deposited on 34 mm glass plates and covered with 20 µg·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' [25] and one above ground in the radionuclide metrology laboratory at JRC-Geel. Point-like sources for gamma measurements were prepared gravimetrically by drop deposition on laminated plastic foils with a diameter of 34 mm. They were measured above ground using two coaxial 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 measurements (ULGS) in HADES using the Ge-8 detector. The Ge-8 is a HPGe detector of type Broad Energy Germanium Detector (BEGe) with a relative efficiency of 20 % (Canberra). The activities of ²⁴¹Am and ²⁴³Am were calculated based on the main gamma-ray peaks of the nuclides, i.e. the 59.54 keV line for ²⁴¹Am and the 74.66 keV line for ²⁴³Am, respectively [26, 27, 28].

4. Results and discussion

4.1 Results of the characterisation of ²⁴¹Pu spike solution

The results of the characterisation of the purified ²⁴¹Pu solution are summarised in Table 1.

		Value	Uncertainty ($k=2$)
Amount content	²⁴¹ Pu [mol·g ⁻¹]	$8.1394 \cdot 10^{-8}$	$0.0053 \cdot 10^{-8}$
Isotope amount fractions	$n(^{238}\text{Pu})/n(\text{Pu}) \cdot 100$	0.00097	0.00034
	$n(^{239}\text{Pu})/n(\text{Pu}) \cdot 100$	0.00053	0.00018
	$n(^{240}\text{Pu})/n(\text{Pu}) \cdot 100$	0.25741	0.00029
	$n(^{241}\text{Pu})/n(\text{Pu}) \cdot 100$	99.29993	0.00052
	$n(^{242}\text{Pu})/n(\text{Pu}) \cdot 100$	0.44116	0.00025

Table 1: Results of the characterisation of the purified ²⁴¹Pu solution

The values of the plutonium isotope amount fractions were found to be in agreement with the values from the certificate of the ²⁴¹Pu material from 1991. The value for the ²⁴¹Pu amount content was calculated from the mean of the 10 blends measured by IDMS. Each blend was measured in replicates on the Triton TIMS using the total evaporation method. The mass fractionation correction was based on the measurement of the IRMM-290/A3 Pu isotopic standard. The results of the IDMS measurements of the ²⁴¹Pu amount in the spike solution are shown in Figure 3.

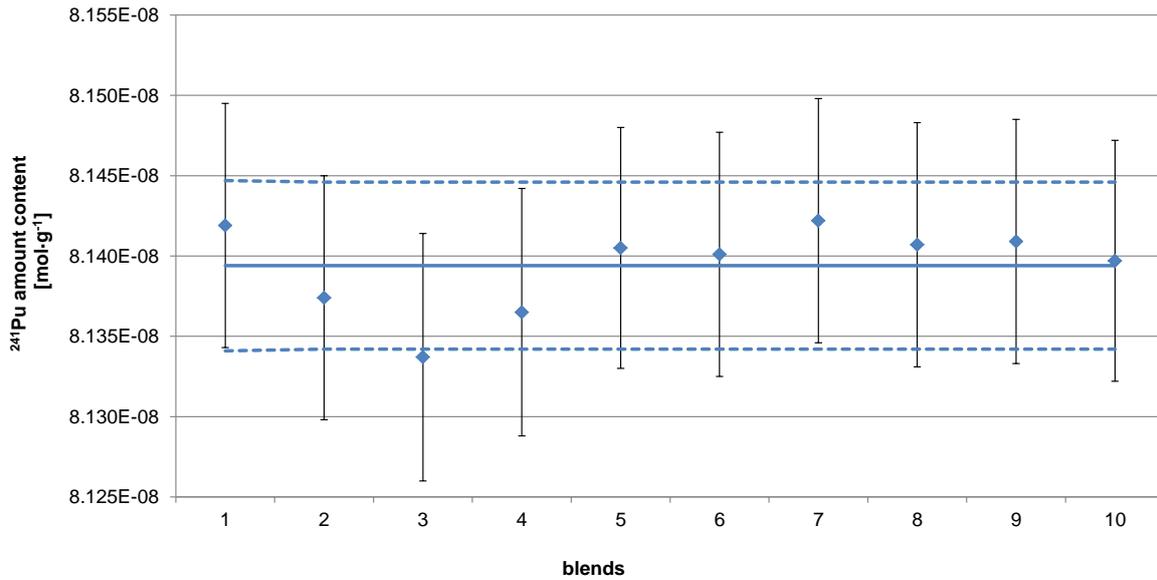


Figure 3: ^{241}Pu amount content in the purified ^{241}Pu solution established by IDMS of 10 blend solutions. The solid line represents the mean of the ten values, the dotted lines represent the expanded uncertainty (coverage factor, $k = 2$) of the mean.

4.2 Results of the verification measurements

Due to the on-going ILC exercise, only the normalised certified value is presented in this paper. The results of the verification measurements obtained by gamma-ray and alpha-particle spectrometry are shown in Figure 4. They are in agreement with the certified values within measurement uncertainty. Measurement uncertainties were estimated according to GUM [29] and mean values calculated by means of the power-moderated mean formalism [30].

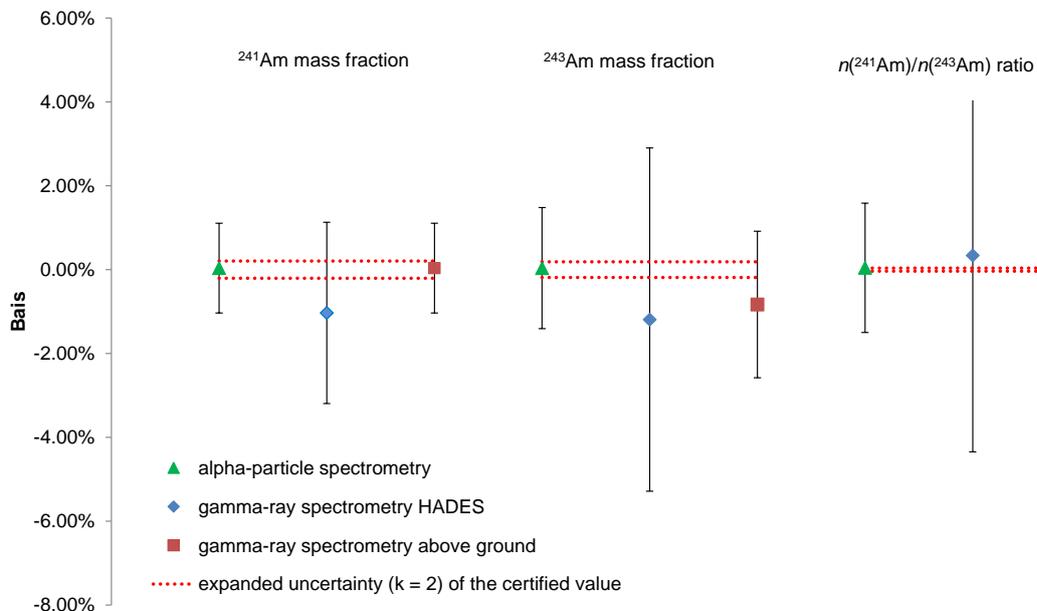


Figure 4: Results of the independent alpha-particle (triangles) and gamma-ray (diamonds and squares) measurements for the ^{241}Am and ^{243}Am mass fractions and $n(^{241}\text{Am})/n(^{243}\text{Am})$ amount ratios expressed as relative differences from the certified value. All the measurement results are shown with

a relative expanded uncertainty ($k = 2$). Red dotted lines show the relative expanded uncertainty ($k = 2$) of the respective certified value.

The relative expanded uncertainty ($k = 2$) for the ^{243}Am amount content from the characterisation assessment was 0.20 %. The major sources of uncertainty come from the half-life of ^{241}Pu , the time of the ^{241}Am ingrowth and the amount content of the ^{241}Pu solution. The relative expanded uncertainties ($k = 2$) of the $n(^{241}\text{Am})/n(^{243}\text{Am})$ and $n(^{242\text{m}}\text{Am})/n(^{243}\text{Am})$ isotope ratios are 0.04 % and 1.0 % respectively.

5. Results and discussion

A novel ^{243}Am spike CRM was prepared and certified for the amount content and isotope amount ratios. The material was produced in compliance with international guidelines. Certified values for the amount content and isotope amount ratios were established by IDMS and TIMS and confirmed by independent radioactivity measurements. The uncertainties associated with the certified values are fit for the purpose for this reference material. This material is available in dilute nitric acid solution with an americium concentration of about 5 ppm.

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