Absolute and relative measurement of the ^{243}Am half-life

M. Marouli¹ · S. Pommé¹ · V. Jobbágy¹ · H. Stroh¹ · R. Van Ammel¹ · A. Fankhauser¹ · R. Jakopič¹ · S. Richter¹ · Y. Aregbe¹ · M. Crozet² · C. Maillard² · C. Rivier² · D. Roudil²

Received: 16 September 2020 / Accepted: 4 October 2020 / Published online: 17 October 2020
© The Author(s) 2020

Abstract

The half-life of ^{243}Am has been measured by an absolute and a relative method, i.e. by determining the specific activity of ^{243}Am and the specific activity ratio with ^{241}Am . A mixed $^{241,243}\text{Am}$ reference material was produced and certified for its americium mass content and its isotope amount ratios. The characterisation of the mass content of ^{243}Am was established by isotope dilution mass spectrometry using an ^{241}Am spike, produced from highly enriched ^{241}Pu material. The isotope amount ratios $n(^{241}\text{Am})/n(^{243}\text{Am})$ and $n(^{242\text{m}}\text{Am})/n(^{243}\text{Am})$ were measured by thermal ionisation mass spectrometry. Activity measurements were performed by alpha-particle counting at a defined solid angle, as well as high-resolution alpha-particle spectrometry. From the $^{243}\text{Am}/^{241}\text{Am}$ activity and isotopic amount ratios, a value of 16.988 (24) was derived for the $^{243}\text{Am}/^{241}\text{Am}$ half-life ratio. Using a value of 432.6 (6) a for the ^{241}Am half-life, the corresponding ^{243}Am half-life value, 7349 (15) a, is in good agreement with the result obtained from the absolute method, 7342 (14) a. The mean value, 7345 (14) a, agrees well with data from literature and lowers the relative standard uncertainty to 0.2%.

Keywords Half-life · ^{243}Am · Nuclear safeguards · Nuclear forensics · Spike reference material · Isotope ratio

Introduction

The decay constant λ establishes the link between the number of atoms N of a particular radionuclide in a sample and their expected radioactive decay rate A , i.e. $\lambda = A/N$ [1]. The value of λ -or inversely the half-life $T_{1/2} = \ln(2)/\lambda$ -is invariable in space and time, in spite of recent claims of variability in decay rate measurements erroneously ascribed to solar influences [2–11]. Accurate values for the decay constant or half-life of trans-uranium nuclides are required for accounting of nuclear material, assay of targets used in nuclear physics experiments, and characterisation of isotopic mixtures for radioanalytical purposes. Whereas mass spectrometry is the most accurate measurement method to determine isotope ratios in nuclear material, also relative

activity measurements can be applied with good accuracy provided that the ratio of the decay constants of the isotopes are well known.

A coordinated research project of the IAEA [12] has identified a need for more accurate decay data for ^{243}Am , comprising the half-life value as well as the alpha and gamma emission characteristics. The longest-lived and most common isotopes of americium, ^{241}Am and ^{243}Am , have half-lives of 432.6 (6) a and 7367 (23) a, respectively [13]. Both are alpha emitters. Whereas any primordial americium on Earth has decayed away, it is produced in nuclear reactors by multiple neutron captures in ^{239}Pu and subsequent β^- decay of ^{241}Pu and ^{243}Pu . A ton of spent nuclear fuel contains about 100 g of americium, thus representing one of the most troublesome long-term radiotoxicity hazards in nuclear waste. Partitioning and transmutation of actinium and curium are being considered to reduce long-term radiotoxicity and heat load of spent fuel [14]. Americium can also be used in Radioisotope Thermoelectric Generators (RTGs) and Radioisotope Heater Units (RHUs) in space nuclear power systems [15].

Recently, the Joint Research Centre (JRC) and the Commissariat à l'énergie atomique et aux énergies alternatives (CEA) have collaborated in the production and certification

✉ S. Pommé
stefaan.pomme@ec.europa.eu

¹ European Commission, Joint Research Centre (JRC), Geel, Belgium

² Commissariat à l'Énergie Atomique Et Aux Énergies Alternatives (CEA), DES, ISEC, DMRC, University of Montpellier, Marcoule, France

of an ^{243}Am reference material for the quantification of Am in an unknown sample by isotope dilution mass spectrometry (IDMS) [16–19]. This responds to a pressing need in the fields of nuclear safeguards and security, nuclear forensics, and accountability in the nuclear fuel cycle [20, 21]. The reference material contains a mixture of ^{241}Am and ^{243}Am , which has been carefully analysed for the amount content by IDMS and for isotope amount ratios by thermal ionisation mass spectrometry (TIMS). In addition, absolute $^{243}\text{Am} + ^{241}\text{Am}$ and relative $^{243}\text{Am}/^{241}\text{Am}$ radioactivity measurements were performed as a redundancy check of the certified amount values.

In this work, the isotope amount content and activity measurements are used to derive a value for the ^{243}Am half-life. A comparison is made with previously published measurement results [23–31] and evaluated values by the Decay Data Evaluation Project (DDEP) and Nuclear Data Sheets (NDS) [13, 32]. Figure 1 shows a schematic overview of the techniques used and the observables obtained through a relative and absolute method, as explained further on in the paper. All uncertainty values in this work pertain to the standard uncertainty.

Fig. 1 Scheme of the methodology. The relative method derives the half-life ratio between ^{243}Am and ^{241}Am from the isotopic abundance ratio of a mixed material via TIMS and the activity ratio via alpha spectrometry. A literature value of $T_{1/2}(^{241}\text{Am})$ is then used to calculate $T_{1/2}(^{243}\text{Am})$. The absolute method uses additional data of the americium content determined via IDMS and the absolute activity of ^{243}Am and ^{241}Am , to determine the specific activity of ^{243}Am

Measurement methods

Material production

The starting solution, containing four milligram americium source material with mass fractions of 88% ^{243}Am and 12% ^{241}Am , was made available by the CEA/L2AT (ATalante Analysis Laboratory). The solution was passed through TRU resin (Triskem, France) to remove the impurities, and the americium was then eluted with 5 mL 0.01 M nitric solution. The purified americium solution was shipped to JRC-Geel and diluted upon arrival in 2400 mL 1 M nitric acid solution to obtain an Am mass fraction of $1.5 \mu\text{g (Am) g}^{-1}$ (solution). This Am fraction 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-cleaned screw-cap ampoules. In total, 587 units were prepared, each unit containing approximately $5 \mu\text{g}$ of americium in 1 M nitric acid solution [16, 19]. Out of the produced units, eighteen were selected for the characterisation of the material.

Amount content by IDMS

The characterisation of the mass content of ^{243}Am was established by IDMS as a primary measurement method. In IDMS, the amount of an element in the sample is determined on the basis of additions of known amounts of the same element with different isotopic composition (called spike). By measuring the change in isotopic composition of the blend (sample-spike mixture) by mass spectrometry, the unknown amount of the element in the sample can be calculated. In the absence of a suitable ^{241}Am spike CRM for IDMS measurements, an alternative approach was applied. An ^{241}Am spike was produced from highly enriched ^{241}Pu ($m(^{241}\text{Pu})/m(\text{Pu}) = 0.993$) material available at JRC-Geel. The purification of approximately 2 mg ^{241}Pu material was done by anion exchange separation. The ingrown ^{241}Am , produced by beta-decay of ^{241}Pu , was used as spike for the measurement of the amount content of ^{243}Am by IDMS. An aliquot of about 2.5 g was taken out of each of the 18 units and mixed with an aliquot of about 1.0 g of the ^{241}Pu spike solution.

Isotope ratios by TIMS

The isotope amount ratios $n(^{241}\text{Am})/n(^{243}\text{Am})$ and $n(^{242\text{m}}\text{Am})/n(^{243}\text{Am})$ of the unspiked material, sampled in

aliquots of 0.7 g, were measured by thermal ionisation mass spectrometry. All isotope ratio measurements were performed using the total evaporation method on a multi-collector Triton TIMS [33].

Activity ratios by alpha spectrometry

High-resolution alpha particle spectrometry [34, 35] was used to determine the activity ratio $A(^{243}\text{Am})/A(^{241}\text{Am})$. The set-up, which is often used for determining alpha-emission probabilities, has been described before [36]. The alpha source was prepared from the Am CRM solution by electrodeposition on a polished stainless steel disk with an active diameter of 18.6 mm. It was measured using passivated ion-implanted planar silicon detectors (PIPS®, Mirion Technologies (MGPI) SA, France).

For the production of the source, an 0.2 cm^3 aliquot of the initial solution was transferred into a 100 cm^3 glass beaker and evaporated to dryness. The residue was dissolved in 10 cm^3 saturated ammonium nitrate (NH_4NO_3) solution (1920 g dm^{-3} ; $T = 20\text{ }^\circ\text{C}$) and transferred into the electrodeposition cell. The beaker was rinsed with 2 cm^3 portions of saturated NH_4NO_3 and finally with 2 cm^3 of deionised water. The total volume of the electrolyte solution was approximately 16 cm^3 , having a $\text{pH} = 3.0$ (1). A mirror-polished stainless steel disk was used as source backing (cathode). A spirally wound platinum wire was used as anode and the cathode-to-anode distance was adjusted to approximately 7 mm. The electrodeposition current was kept stable at 0.85 (3) A at a voltage between 4.6 and 5 V over a period of 75 min. One minute before switching off the current, 1 cm^3 of 25% ammonium hydroxide (NH_4OH) was added to the solution to prevent the re-dissolution of the deposited americium layer. The source was rinsed with deionised water and dried at ambient temperature.

The source was measured at 3.6 cm source-to-detector distance using two PIPS® detectors of 150 mm^2 and 300 mm^2 active area. A magnet system on top of the source was used to deflect the conversion electrons away from the detector [37]. The intermediate spectra were acquired in daily cycles and shifted prior to summing into a composite spectrum, to compensate for any electronic gain drift during the measurement campaign [38, 39]. Three composite spectra were obtained and analysed with the deconvolution software 'BEST' [40]. The number of counts in the ^{241}Am and ^{243}Am peaks were between 3×10^6 and 2×10^7 . As an example, the measured and fitted alpha-particle energy spectra obtained with the 300 mm^2 PIPS® detector are shown in Fig. 2. The attained energy resolution was approximately 12.2 keV (FWHM) with the 150 mm^2 detector and 15.8 keV (FWHM) with the 300 mm^2 detector.

Fig. 2 Measured (dots) and fitted (lines) alpha spectrum of the $^{243,241}\text{Am}$ material, with interference peaks from ^{238}Pu and tailing from other nuclides at higher energy (bottom). Residuals to the fit (top)

Activity by alpha counting

The total Am activity per unit mass was determined by means of alpha-particle counting at a defined solid angle (DSA) [36, 41, 42]. This is potentially the most accurate primary standardisation technique for the activity of alpha-emitting nuclides [42]. By measuring a small fraction of alpha-particles emitted perpendicularly from the source plane, loss of particles by absorption in the source material at low emission angles is avoided. The detection efficiency practically equals the geometrical efficiency, which can be calculated from solid-angle formulas [43–45]. The accuracy of the method depends particularly on the reproducibility and accurate knowledge of the geometrical conditions. It is further improved by taking autoradiographs of the source activity, to tailor the solid-angle calculation to the radial activity distribution over the source area [42, 45].

Gravimetrically quantified drops of americium solution were deposited on 34-mm-diameter glass plates, with active areas of less than 19 mm diameter. They were dried on a heat plate at $110\text{ }^\circ\text{C}$ and covered with $20\text{ }\mu\text{g cm}^{-2}$ VYNS foils (polyvinylchloride-polyvinylacetate copolymer) to prevent material loss. The sources were measured in two set-ups [36] with large passivated implanted planar silicon detectors of 3000 mm^2 and 5000 mm^2 surface area, at 5.7% and 6.7% geometrical efficiency ($=\text{solid angle}/4\pi$), respectively. The source-to-detector distances were accurately measured using both an optical focusing measuring device and a 3D coordinate measurement machine [46]. The energy threshold was set at approximately 1.5 MeV, thus avoiding detection of β -particles from the decay of ^{239}Np . The total count rate ranged between 35 s^{-1} and 120 s^{-1} , whereas the background rate was less than 0.03 s^{-1} . Dedicated lifetime clock gates were used in combination with artificially imposed dead time [47]. The mean dead

time was less than 0.07% and the cascade effect with pulse pileup [48] was insignificant (<0.001%).

Analysis and results

Mass metrology

The mass metrology and mass spectrometry work performed in the frame of the characterisation of the americium reference material has been described elsewhere [16–18]. The resulting absolute amount contents and relative isotope fractions are provided in Table 1. The values are confirmed by an interlaboratory comparison organised by CEA/CETAMA using IRMM-0243/STAM as the test sample in the context of the certification of the ^{243}Am spike [19].

Relative activity method

In the relative activity method, the half-life of ^{243}Am is determined relatively to the half-life of ^{241}Am , $T_{1/2}(^{241}\text{Am})=432.6$ (6) a [13], which is known with a relative standard uncertainty of 0.14%. From the definition of the decay constant follows a simple relationship for the $^{243}\text{Am}/^{241}\text{Am}$ half-life ratio

$$\frac{T_{1/2}^{243}}{T_{1/2}^{241}} = \frac{\lambda_{241}}{\lambda_{243}} = \frac{A_{241}}{A_{243}} \frac{N_{243}}{N_{241}} \quad (1)$$

in which A_{241}/A_{243} represents the $^{241}\text{Am}/^{243}\text{Am}$ activity ratio and N_{243}/N_{241} the $^{243}\text{Am}/^{241}\text{Am}$ amount ratio in the

americium material at the reference date. The amount ratio equals the mass fraction ratio divided by the atomic weight ratio.

The activity ratios derived from the three acquired alpha spectra are presented in Table 2. The number of counts C in the fitted peaks are corrected for decay during the measurement. The count rate is extrapolated to the reference date via the nuclide-specific f_d decay correction factor [1]

$$f_d = \frac{\lambda t_m}{(1 - e^{-\lambda t_m})} e^{\lambda(t-t_{ref})} \quad (2)$$

in which t_m is the duration of the measurement, $(t - t_{ref})$ is the time difference between the start of the measurement and the reference time, and λ is the decay constant of either ^{241}Am or ^{243}Am , such that the activity ratio at reference time is

$$\frac{A_{241}}{A_{243}} = \frac{C_{241}}{C_{243}} \frac{f_d^{241}}{f_d^{243}} \quad (3)$$

The ratios of the decay correction factors deviated by no more than 0.15% from unity, and their relative uncertainties were negligibly small.

As for the uncertainty estimate, besides the Poisson statistics governing the peak areas, there is an important uncertainty component to be considered from the tailing of the ^{241}Am peak underneath the ^{243}Am peak area. Wrong assessment of the tailing area leads to a fully correlated error in the nominator and the denominator of the activity ratio. The corresponding variance is propagated by a factor of 4, as explained in the case

Table 1 Measured amount contents and isotopic amount ratios in the reference material IRMM-0243 ($^{243,241}\text{Am}$ in 1 M HNO_3 solution) measured by IDMS and TIMS [16–19] and their standard uncertainties at the reference date 1 January 2017

Entity	Unit	^{241}Am	^{243}Am	Am	^{242m}Am
Amount content	nmol/g (solution)	0.7754 (8)	5.696 (6)	6.472 (6)	–
Mass fraction (%)	$\mu\text{g/g}$ (solution)	0.18692 (18)	1.3845 (13)	1.5716 (15)	–
Isotope amount fraction (%)	mol/mol (Am)	11.9810 (21)	88.0069 (21)		0.01208 (11)*

*Indicative value derived from the interlaboratory comparison [19]

Table 2 Number of thousands of counts in the ^{241}Am and ^{243}Am peaks derived from the deconvolution of three composite alpha-particle spectra. Correction factors for decay (uncertainty <0.001%),

to convert count rates into activity at the reference date. Derived $^{241}\text{Am}/^{243}\text{Am}$ activity ratio and statistical uncertainty

Spectrum	^{241}Am		^{243}Am		Activity ratio $^{241}\text{Am}/^{243}\text{Am}$
	Counts (10^3)	Decay correction	Counts (10^3)	Decay correction	
1	2290.2 (15)	0.999425	988.7 (16)	0.999966	2.315 (5)
2	2728.3 (16)	0.999941	1179.9 (12)	0.999997	2.312 (4)
3	19,235.4 (43)	1.001629	8330.7 (31)	1.000096	2.313 (1)
Mean	–	–	–	–	2.313 (1)

study I in Ref. [35]. To account for additional correlations, a model of the experiment was set up and the GUM Workbench software [49] was used to estimate the uncertainties for the counts C_{241} , C_{243} and the relative activity A_{241}/A_{243} , similar to the method used in Ref. [50]. It is compliant with the 'ISO Guide to the Expression of Uncertainty in Measurement' [51] and the requirement document EA 4/02 of the European Cooperation for Accreditation [52].

The spectrum was hypothetically divided into two energy regions, region 'A' [4700–5360] keV and region 'B' [5361–5580] keV, which mainly cover the ^{243}Am and ^{241}Am peak areas and contain C_A and C_B counts, respectively. The regions contain additional counts, $C_{A,t}$ and $C_{B,t}$, mainly originating from the tailing of other alpha emitters having peaks at higher energy, such as ^{244}Cm and ^{242}Cm . The area of the tailing part of the ^{241}Am peak interfering in region A is denoted $C_{B,A}$. The model equations to investigate the uncertainties on the Am peak areas C_{241} and C_{243} are

$$\begin{cases} C_{243} = C_A - C_{A,t} + C_{A,f} \\ C_{241} = C_B - C_{B,t} + C_{B,A} + C_{B,f} \end{cases} \quad (4)$$

Two additional components, $C_{A,f}$ and $C_{B,f}$, represent a measure for the goodness of the fit. Their nominal value is set equal to zero, but their uncertainty is taken as the sum of the absolute residuals, i.e. the difference between measurement and fit in each spectrum channel. For the other components, the square root of the number of counts is taken as the statistical uncertainty value. The mean activity ratio was determined as $A_{241}/A_{243} = 2.313$ (1) (see Table 1), and the half-life ratio $T_{1/2}^{243}/T_{1/2}^{241} = 16.988$ (24). With 0.14% relative standard uncertainty on the ^{241}Am half-life as well as on the half-life ratio, the ^{243}Am half-life was determined with 0.2% relative uncertainty as 7349 (15) a.

Specific activity method

By measuring the activity of the ^{243}Am with an absolute method, the dependence on the ^{241}Am half-life can in principle be eliminated. In this experiment, no pure ^{243}Am material was available. Therefore, the total activity $A_{\text{tot}} = A_{241} + A_{243}$ was measured and the contribution of ^{241}Am was subtracted on the basis of the relative activity A_{241}/A_{243} . Consequently, the measurement results of the absolute and relative methods in this work are correlated to some extent, so that the 'absolute' method de facto qualifies as a 'hybrid' method.

The measurement equation follows directly from $A_{243} = \lambda_{243}N_{243}$

$$T_{1/2}^{243} = \ln(2) \frac{N_{243}}{m} \frac{m}{A_{\text{tot}}} \left(1 + \frac{A_{241}}{A_{243}} \right) \quad (5)$$

in which N_{243}/m and A_{tot}/m are the number of ^{243}Am atoms and the $^{241}\text{Am} + ^{243}\text{Am}$ activity per gram of solution, respectively.

The activity at the reference date, $A_{\text{tot}}(t_{\text{ref}})$ (Bq), is obtained by counting alpha particles under a small but well-defined solid angle, using the equation

$$A_{\text{tot}} = \frac{C_{\text{tot}}}{t_m} \frac{4\pi}{\Omega} f_{\text{tail}} f_d \quad (6)$$

in which C_{tot}/t_m is the number of counts per system live-time, which is adequately corrected for count loss through pileup and system dead time by imposing a well-known dead time for each registered pulse [47, 48]; Ω is the solid angle in steradian [41, 43–45]; f_{tail} is a correction factor for the extrapolation of the spectrum below the imposed low-energy threshold [36]; and f_d is a weighted decay correction factor (Eq. 2) for both Am isotopes, based on the activity ratio derived from alpha spectrometry.

The total number of counts C_{tot} had to be corrected for the presence of radioactive impurities. In Fig. 3, decay schemes are shown for $^{241,243,242\text{m}}\text{Am}$ and ^{244}Cm , in view of their relevance to this part of the work. Interference from the decay products of $^{241,243}\text{Am}$ is considered negligible. With high-resolution alpha-particle spectrometry (see relative method), alpha peaks from the decay of ^{244}Cm and $^{242\text{m}}\text{Am}$ were identified. Small alpha peaks from ^{238}Pu fed through the decay of ^{242}Cm could not be discerned because they were masked by the ^{241}Am peaks. Assuming that the material was free from Pu at the last chemical purification on 28 April 2015 [16–18], the ingrowth of ^{238}Pu was calculated as approximately 0.8% of the ^{242}Cm activity at the reference date. The contribution of the impurities to the total activity was measured as 0.19 (2) %. This fraction is consistent with the value 0.21 (1) % derived from the interlaboratory comparison [19].

An overview of the uncertainty assessment of a typical activity measurement is presented in Table 3. Corrections for decay during the short measurement period t_m were insignificant. Half of the tailing correction f_{tail} is taken as its uncertainty. Backscattering effects of alpha-particles on the glass plate and self-absorption of the material are possible, but with less than 0.02% probability each, and are introduced into the uncertainty budget. The activity concentration values of the solution calculated via Eq. 6 for each source are presented in Table 4. All results were internally consistent.

With 0.16% relative standard uncertainty on the $^{241}\text{Am} + ^{243}\text{Am}$ activity per unit mass, 33.99 (6) Bq/g, and 0.14% on the activity ratio, 16.988 (24), the ^{243}Am half-life was determined with 0.2% relative uncertainty as 7342 (14) a.

Fig. 3 Decay chains of the main radionuclides present in the radioactive solution

Discussion

The results from both methods agree extremely well, and their average value 7345 (14) a is taken as the final estimate of the ²⁴³Am half-life. There is significant correlation between the two methods, both relying on the measured activity ratio between the isotopes. Therefore, the uncertainty value of the more precise method was adopted directly to the mean half-life instead of the combined uncertainty, since a rigorous error propagation would not result in a significantly lower uncertainty. The results obtained in this work are consistent with literature values. An overview of published experimental half-life values is presented in

Table 5 [13] and shown graphically in Fig. 4. The half-life ratio agrees within one combined standard uncertainty with the five other published data. Also the specific activity measurements are mutually compatible within one combined standard uncertainty. The uncertainties in this work are lower, mainly due to the more accurately performed alpha-particle counting and spectrometry measurements.

A power-moderated mean (PMM) with $\alpha = 1$ – where the number α denotes the power of the uncertainties in the weighting factors [53] – of the three most recent (and consistent) half-life ratio data in Table 5 results in 17.011 (28) a. This is compatible with, but less precise than the value 16.988 (24) measured in this work, or the slightly lower

Table 3 Typical standard uncertainty budget of the absolute activity concentration of the solution derived from measuring a source by alpha-particle counting at a defined solid angle

Component	Relative uncertainty (%)
Counting statistics	0.01
Dead time	<0.01
Pile-up	<0.001
Background	<0.001
Geometric efficiency	0.10
Low-energy tailing	0.02
Scattering at detector	0.02
Self-absorption	0.02
Impurities	0.02
Weighing	0.12
Total	0.16

Table 4 Total activity per gram of solution at reference date 1 January 2017 as calculated from the measurements of the individual sources standardised by alpha-particle counting at a defined solid angle. The relative standard uncertainty on the $^{243}\text{Am} + ^{241}\text{Am}$ activity is 0.16%

Source	Activity (kBq/g)*	
	DSA1 set-up	DSA2 set-up
Am243G1604	34.15 (9)	34.04 (10)
Am243G1605	34.15 (6)	34.05 (6)
Am243G1606	34.06 (6)	34.06 (6)
Am243G1607	34.09 (6)	34.05 (6)
Am243G1609	34.11 (5)	34.05 (5)
Am243G1610	34.02 (5)	34.04 (7)
Am243G1611	34.02 (5)	34.01 (6)
Am243G1612	34.05 (6)	34.04 (6)
Average	34.06 (5)	–
$^{243}\text{Am} + ^{241}\text{Am}$	33.99 (6)	–

*Including impurity fraction of 0.19 (2) %

value 16.980 (24) derived from the mean half-life result 7345 (14) a from both methods. The PMM ($\alpha = 2$) of the latter with the three literature data yields 16.998 (17), which can be recommended as a good overall estimate. This ratio in combination with $T_{1/2}(^{241}\text{Am}) = 432.6$ (6) a leads to a recommended half-life value of $T_{1/2}(^{243}\text{Am}) = 7353$ (10) a.

Conclusions

The ^{243}Am half-life and the $^{243}\text{Am}/^{241}\text{Am}$ half-life ratio have been measured accurately by combining mass spectrometry through IDMS and TIMS, high-resolution alpha-particle spectrometry, and alpha-particle counting measurements.

Table 5 Overview of measured ^{243}Am half-life values, determined by specific activity or relative to the ^{241}Am half-life ($= 432.6$ (6) a) and the recommended evaluated value by the DDEP [13]. Below are the half-life values obtained in this work and their mean value

Year	Method	$T_{1/2}^{243}/T_{1/2}^{241}$	$T_{1/2}^{243}\text{(a)}$
1959	Relative activity	16.85 (35)	7289.3 (1517)
1960	Relative activity	16.70 (10)	7224.4 (1000)
1968	Relative activity	16.96 (13)	7336.9 (572)
1968	Specific activity	–	7390 (50)
1974	Specific + Relative activity	–	7380 (34)
1980	Relative activity	17.010 (95)	7359 (42)
2007	Relative activity	17.022 (27)	7363.7 (230)
DDEP	–	–	7367 (23)
2020	Relative activity	16.988 (24)	7349 (15)
2020	Specific activity (hybrid)	–	7342 (14)
Mean	–	16.980 (24)*	7345 (14)

*Calculated from the mean ^{243}Am half-life obtained with both methods

Fig. 4 Comparison of previously measured ^{243}Am half-life values (Table 5) with the ‘New’ result from this work and a recommended ‘Mean’ value calculated from the four most recent $^{243}\text{Am}/^{241}\text{Am}$ half-life ratios

The values derived from a mean of two methods, 7345 (14) a for the ^{243}Am half-life and 16.980 (24) for the $^{243}\text{Am}/^{241}\text{Am}$ half-life ratio, have the smallest uncertainty hitherto reported, yet agree very well with published experimental work. In combination with three consistent literature data, and assuming that $T_{1/2}(^{241}\text{Am}) = 432.6$ (6) a, mean values of $T_{1/2}(^{243}\text{Am})/T_{1/2}(^{241}\text{Am}) = 16.998$ (17) and $T_{1/2}(^{243}\text{Am}) = 7353$ (10) a can be recommended as good estimates.

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons licence, and indicate if changes were made. The images or other third party material in this article are

included in the article's Creative Commons licence, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons licence and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this licence, visit <http://creativecommons.org/licenses/by/4.0/>.

References

- Pommé S (2015) The uncertainty of the half-life. *Metrologia* 52:S51–S65
- Pommé S et al (2016) Evidence against solar influence on nuclear decay constants. *Phys Lett B* 761:281–286
- Pommé S et al (2017) On decay constants and orbital distance to the Sun—part I: alpha decay. *Metrologia* 54:1–18
- Pommé S et al (2017) On decay constants and orbital distance to the Sun—part II: beta minus decay. *Metrologia* 54:19–35
- Pommé S et al (2017) On decay constants and orbital distance to the Sun—part III: beta plus and electron capture decay. *Metrologia* 54:36–50
- Pommé S et al (2018) Is decay constant? *Appl Radiat Isot* 134:6–12
- Pommé S, Kossert K, Nähle O (2017) On the claim of modulations in ^{36}Cl beta decay and their association with solar rotation. *Solar Phys* 292:162
- Pommé S, Lutter G, Marouli M, Kossert K, Nähle O (2017) On the claim of modulations in radon decay and their association with solar rotation. *Astropart Phys* 97:38–45
- Pommé S, Lutter G, Marouli M, Kossert K, Nähle O (2018) A reply to the rebuttal by Sturrock et al. *Astropart Phys* 107:22–25
- Pommé S (2019) Solar influence on radon decay rates: irradiance or neutrinos? *Eur Phys J C* 79:73
- Pommé S, Pelczar K (2020) On the claim of correlation between radioactive decay rates and space weather. *Eur Phys J C* (in press)
- Kellett M (2012) Assessment of actinide decay data evaluations: findings of an IAEA coordinated research project. *Appl Radiat Isot* 70:1919–1923
- Decay Data Evaluation Project (2004–2020) DDEP Monographie BIPM-5—Table of Radionuclides <https://www.lnhb.fr/nuclear-data/nuclear-data-table/> Accessed 4 Aug 2020
- Kooyman T, Buiron L, Rimpault G (2018) A comparison of curium, neptunium and americium transmutation feasibility. *Ann Nucl Energy* 112:748–758
- Ambrosi RM et al (2019) European radioisotope thermoelectric generators (RTGs) and radioisotope heater units (RHUs) for space science and exploration. *Space Sci Rev* 215:55
- Jakopič R et al (2017) ^{243}Am certified reference material for nuclear safeguards and security. Proceedings 39th ESARDA Annual Meeting—Symposium, 16–18 May 2017, Düsseldorf, Germany, pp 409–421
- Jakopič R et al (2017) Preparation and certification of ^{243}Am spike reference material: IRMM-0243, EUR 28748 EN
- Jakopič R et al (2020) ^{243}Am certified reference material for mass spectrometry. *JRNC* (in press)
- Crozet M et al (2019) Contribution of an interlaboratory comparison to the certification of the STAM/IRMM-0243 ^{243}Am reference material. *J Radioanal Nucl Chem* 319:717–725
- Inn K et al (2013) The urgent requirement for new radioanalytical certified reference materials for nuclear safeguards, forensics, and consequence management. *J Radioanal Nucl Chem* 296:5–22
- Aggarwal SK (2016) Nuclear forensics: what, why and how? *Curr Sci* 110:782–791
- Aggarwal SK (2016) Thermal ionisation mass spectrometry (TIMS) in nuclear science and technology – a review. *Anal Meth* 8:942–957
- Diamond H, Fields PR, Mech J, Inghram MG, Hess DC (1953) The half-life of Am^{243} . *Phys Rev* 92:1490–1491
- Asaro F, Perlman I (1954) Decay properties of Am^{243} and possible rotational bands in the alpha spectra of odd-even nuclei. *Phys Rev* 93:1423–1424
- Wallmann JC, Graf P, Goda L (1958) The specific activities and half lives of americium-241 and americium-243. *J Inorg Nucl Chem* 7:199–200
- Barnes RF, Henderson DJ, Harkness AL, Diamond H (1959) The alpha and electron capture partial half-lives of ^{242}Am . *J Inorg Nucl Chem* 9:105–107
- Beadle AB, Dance DF, Glover KM, Milsted J (1960) The half-life of americium-243. *J Inorg Nucl Chem* 12:359–361
- Brown LC, Propst RC (1968) A new determination of the half-life of ^{243}Am . *J Inorg Nucl Chem* 30:2591–2594
- Polyukhov VG, Timofeev GA, Privalova PA, Gabeskiriya VY, Chetverikov AP (1974) Determination of ^{243}Am half-life. *At Energy* 37:357–359 (1974); *Sov At Energy* 37:1103–1105 (1975)
- Aggarwal SK, Parab AR, Jain HC (1980) Half-life of ^{243}Am . *Phys Rev C* 22:767–773
- Aggarwal SK, Alamelu D, Shah PM, Mirashi NN (2007) Precise and accurate determination of the half-life of the α -decay nuclide ^{243}Am using ^{241}Am as the reference isotope. *Nucl Instr Meth A* 571:663–668
- Nesaraja CD, McCutchan EA (2014) Nuclear Data Sheets for $A = 243$. *Nucl Data Sheets* 121:695–748
- 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
- García-Torano E (2006) Current status of alpha-particle spectrometry. *Appl Radiat Isot* 64:1273–1280
- Pommé S (2015) Typical uncertainties in alpha-particle spectrometry. *Metrologia* 52:S146–S155
- Pommé S, Sibbens G (2008) Alpha-particle counting and spectrometry in a primary standardisation laboratory. *Acta Chim Slov* 52:111–119
- Paepen J, Dirican A, Marouli M, Pommé S, Van Ammel R, Stroh H (2014) A magnet system for the suppression of conversion electrons in alpha spectrometry. *Appl Radiat Isot* 87:320–324
- Pommé S, Sibbens G (2004) Concept for an off-line gain stabilisation method. *Appl Radiat Isot* 60:151–154
- Pommé S, Sibbens G (2004) A new off-line gain stabilisation method applied to alpha-particle spectrometry. In: Ciarlini P, Cox MG, Pavese F, Rossi GB (eds) *Advanced Mathematical and Computational Tools in Metrology VI, Series on Advances in Mathematics for Applied Sciences*, vol 66. World Scientific Publishing Company, Singapore, pp 327–329
- Pommé S, Marroyo BC (2015) Improved peak shape fitting in alpha spectra. *Appl Radiat Isot* 96:148–153
- Pommé S (2007) Methods for primary standardization of activity. *Metrologia* 44:S17–S26
- Pommé S (2015) The uncertainty of counting at a defined solid angle. *Metrologia* 52:S73–S85
- Pommé S, Johansson L, Sibbens G, Denecke B (2003) An algorithm for the solid angle calculation applied in alpha-particle counting. *Nucl Instrum Methods A* 505:286–289
- Pommé S, Paepen J (2007) A series expansion of Conway's generalised solid-angle formulas. *Nucl Instrum Methods A* 579:272–274

45. Sibbens G, Pommé S, Johansson L, Denecke B (2003) Tailoring solid angle calculations to the actual radioactivity distribution of planar sources. *Nucl Instrum Methods A* 505:277–281
46. Marouli M, Pommé S (2019) Automated optical distance measurements for counting at a defined solid angle. *Appl Radiat Isot* 153:108821
47. Pommé S, Fitzgerald R, Keightley J (2015) Uncertainty of nuclear counting. *Metrologia* 52:S3–S17
48. Pommé S (2008) Cascades of pile-up and dead time. *Appl Rad Isot* 66:941–947
49. MetroData GmbH, GUM Workbench Version 2.4: Software Package for the Evaluation of Uncertainty, <https://www.metrodata.de>
50. García-Toraño E, Crespo T, Marouli M, Jobbágy V, Pommé S, Ivanov P (2019) Alpha-particle emission probabilities of ^{231}Pa derived from first semiconductor spectrometric measurements. *Appl Rad Isot* 154:108863
51. BIPM/ISO: Guide to the Expression of Uncertainty in Measurement, International Organization for Standardization, 1995, ISBN 92-67-20188-3
52. European co-operation for Accreditation: Expression of the uncertainty of measurement in calibration, EA, 1999, EA-4/02
53. Pommé S, Keightley J (2015) Determination of a reference value and its uncertainty through a power-moderated mean. *Metrologia* 52:S200–S212

Publisher's Note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.