Applied Radiation and Isotopes 230 (2026) 112433

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Standardization of ^{239}Pu using isotopic ratios obtained through alpha spectrometry and mass spectrometry

A.J. Pearce *, M.R. Bodine, R. Collé, R. Essex, J. La Rosa, L. Laureano-Pérez, L. Pibida, M.A. Tyra

Physical Measurement Laboratory, National Institute of Standards and Technology, 100 Bureau Dr, Gaithersburg, MD, 20899, USA

ABSTRACT

The standardization of a new ^{239}Pu radioactivity solution with considerable impurity was performed. The massic activity of ^{239}Pu was obtained through 4πα liquid scintillation (LS) counting with correction for significant amounts of other plutonium isotopic impurities made by a combination of isotope dilution alpha spectrometry and isotopic composition measurements by mass spectrometry.

1. Introduction

Plutonium-239 is of special interest for nuclear non-proliferation, safeguards, and environmental monitoring analyses (Moody et al., 2014). The determination of plutonium in complex environmental samples (introduced, e.g., by atmospheric weapons testing, accidental releases from reprocessing facilities and damaged nuclear reactors, and radioactive waste repository leakage) has promoted the development of highly sensitive analytical techniques, which in turn demand well-characterized plutonium reference materials to assess applicability in said analytical techniques to make environmental plutonium determinations, and to ensure accuracy. Due to a variety of factors — including the discontinuation of military plutonium separations by the mid-1990s (Gray et al., 2015) — there has been a dearth of high purity ^{239}Pu available for reference material production, necessitating the standardization of ^{239}Pu containing other radionuclidic impurities. 4πα liquid scintillation counting (LSC) is the primary standardization method used for ^{239}Pu, but it is insensitive to impurities and thus requires knowledge of the relative isotopic ratios to accurately quantify the ^{239}Pu massic activity in non-pristine samples.

Secondary activity measurements are often used for confirmation on primary methods (Dersch, 2004). Though primary standardizations are generally performed on sources containing a single radionuclide, impurities can be corrected for by secondary methods, which provide invaluable resolving information between species in solution (Pommé, 2007). Typical methods include alpha spectrometry and gamma spectrometry, which can better resolve the energy of α-particle and photon emissions, respectively, than LSC methods. Mass spectrometry has emerged as a powerful technique in radionuclide metrology (Braysher et al., 2020; Pommé, 2022), and has been used in the certification of reference materials containing more than one radionuclide of interest (Crozet et al., 2019). When mass spectrometry measurements are combined with activity measurements, such as alpha spectrometry, half-life determinations through specific activity measurements of long-lived radionuclides becomes possible (Bièvre and Verbruggen, 1999; Cassette et al., 2010; Kossert et al., 2013; Marouli et al., 2020; Nedjadi et al., 2012; Parsons-Davis et al., 2018; Varga et al., 2016; Yang et al., 2010).

The National Institute of Standards and Technology (NIST) has issued ^{239}Pu radioactive solution Standard Reference Materials (SRMs) dating back to 1975 with SRM 4330, followed by SRM 4330a (1996), SRM 4330b (2000) and SRM 4330c (2010). Though these issues have been derived from various stock materials, they have universally contained negligible impurities where the sum of all radioactive impurity activities were less than the uncertainty in the activity of the standardized ^{239}Pu activity.

Here, we describe the production of a new ^{239}Pu radioactivity solution standard. Unlike past issues, this solution contains well-characterized but significant amounts of other isotopes of plutonium. We used a combination of secondary standardization methods — alpha spectrometry and mass spectrometry — to obtain an impurity profile determined as isotopic ratios relative to ^{239}Pu to apply a correction to the primary standardization of a^{239}Pu SRM by 4πα LSC. In addition to these techniques, photon-emitting impurities were also monitored by gamma spectrometry.

This article is part of a special issue entitled: ICRM 2025 published in Applied Radiation and Isotopes.

* Corresponding author.

E-mail address: adam.pearce@nist.gov (A.J. Pearce).

https://doi.org/10.1016/j.apradiso.2026.112433
Received 31 July 2025; Received in revised form 20 November 2025; Accepted 9 January 2026
Available online 10 January 2026
0969-8043/Published by Elsevier Ltd.

2. Experimental methods

2.1. Overview of SRM and counting source production

Fig. 1 provides an overview for the production and measurement campaign for SRM 4330d. A solution weighing 5.45517 g of ^{239}Pu (E & Z IPL ^{239}Pu Solution, reported activity: 36.98 kBq; 6.779 kBq g^{-1}) dissolved in 3 mol L^{-1} HNO_3 was obtained for SRM production from Eckert & Ziegler Analytics (EZA). This solution was originally prepared gravimetrically from a stock solution (DAMRI 25661) calibrated by the Commissariat à l'énergie atomique et aux énergies alternatives (CEA), but otherwise the provenance of the solution is unknown. DAMRI 25661 was calibrated by LS counting and the purity was checked by high-purity germanium (HPGe) gamma spectrometry and alpha spectrometry. The stock solution is known to have a sizeable impurity profile; the certificate of calibration reports activities for alpha-emitting impurities ^{241}Am (280 Bq; $A_{241Am}/A_{239Pu} = 0.0076$) and ^{240}Pu (1200 Bq; $A_{240Pu}/A_{239Pu} = 0.032$), and an activity for beta-emitting ^{241}Pu (550 Bq; $A_{241Pu}/A_{239Pu} = 0.015$).

A dilution of the stock solution of ^{239}Pu was performed with 2.15 g of stock solution and 3 mol L^{-1} HNO_3 solution, giving a gravimetric dilution factor of 182.08(4) (all uncertainty values correspond to the standard uncertainty except where $k = 2$ is indicated to express expanded uncertainty) to make 350 mL of SRM Master solution. An aliquot of the stock solution was set aside, and the remainder of the stock, amounting to 2.233 g, was reserved for future SRM production. The SRM Master solution was dispensed in 5 mL aliquots via a Microlab 620 single-syringe automatic dispenser (Hamilton Company, Reno, NV, USA) into 71 NIST-3 ampoules (Collé, 2019). The remaining 2 g of the SRM Master solution was used to partially fill a 72nd ampoule.

Timing for this work was with respect to the SRM 4330d reference date of July 28th, 2024 ($t = 0$).

Because of the high level of impurity reported in the stock solution, a campaign to analyze the impurity profile by several methods was devised: gamma spectrometry to look for photon-emitting impurities, alpha spectrometry to resolve the various alpha-emitting impurities, and mass spectrometry to analyze for small quantities of long-lived impurities and to obtain a reliable activity ratio of the ^{240}Pu activity and the ^{239}Pu activity (A_{240Pu}/A_{239Pu}) that cannot be determined by alpha spectrometry alone due to the alpha energy overlap between the two species.

2.2. Liquid scintillation

Two LS comparison experiments were conducted: LS Comparison 1 and LS Comparison 2. For LS Comparison 1, one ampoule of SRM 4330d was used to prepare LS sources in two different scintillation cocktail compositions — a set in Ultima Gold AB (UGAB) and a set in Hionic-Fluor (HiF). An ampoule from the previous ^{239}Pu SRM (4330c) was also used to prepare sources, allowing for a comparison between 4330c and 4330d in matching LS cocktail compositions to SRM 4330d. Cocktail composition made with the aim of 5 % water fraction included HNO_3, di (2-ethylhexyl)phosphoric acid (HDEHP), scintillant, and SRM addition. Table 1 shows the characteristic cocktail composition across the set of samples. Ten LS sources were prepared from the SRM 4330d ampoule — 5 of UGAB and 5 of HiF. One of the HiF samples (PuDHiF2) had unreliable mass measurements and was discarded from the data set. The 9 samples of 4330d were compared with 10 samples of 4330c, which were also prepared as 5 of UGAB and 5 of HiF. LS Comparison 2 was conducted identically to LS Comparison 1, with the only exception being the ampoules used to source SRMs 4330d and 4330c and the number of samples measured: 12 samples of each SRM were prepared — 6 of UGAB and 6 of HiF. While only two ampoules of SRM 4330d were measured in the LS measurement campaign, ampoule-to-ampoule homogeneity is anticipated for a solution SRM, and any heterogeneity is anticipated to be within uncertainties.

Two different instruments were used for all LS measurements: a Packard TriCarb A2500 TR (PerkinElmer, Wesley, MA, USA) and a Beckman LS 6500 (Beckman Coulter, Fullerton, CA, USA). Details on normal instrument parameters have been described previously (Laureano-Pérez et al., 2007). Measurements for both LS comparison experiments were divided into two runs — the first run of LS Comparison 1 began on July 23rd, 2024 ($t = -5$ d) and ended on July 30th, 2024 ($t = 2$ d), and the second run began on July 30th, 2024, and ended August 5th, 2024 ($t = 8$ d). Measurements for LS Comparison 2, run 1 began on February 24th, 2025 ($t = 211$ d) and ended on February 27th, 2025 ($t = 214$ d), and measurements of run 2 began on February 27th, 2025, and ended on March 6th, 2025 ($t = 221$ d). Each measurement consisted of 5 cycles of 30-min counts.

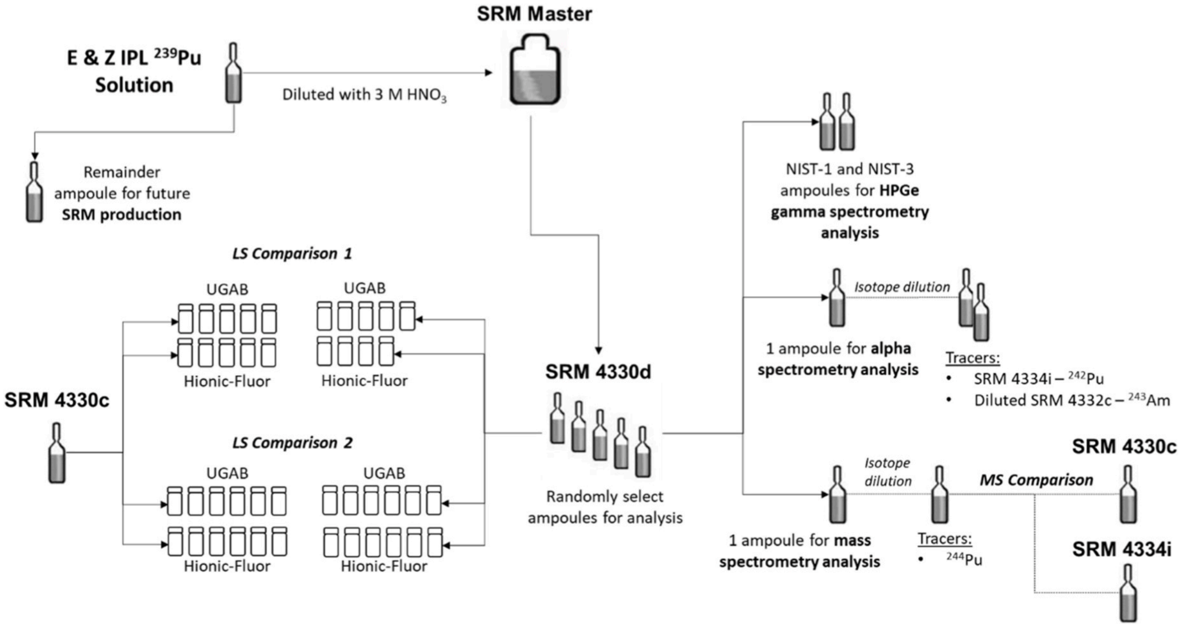

Fig. 1. SRM 4330d production and measurement scheme. Refer to text for details.

Table 1
Composition of LS cocktails used in LS Comparison experiments.

Comparison	Sample	Scintillant	Mass of scintillant (g)	Aliquot mass of HDEHP (g)	Aliquot mass of SRM solution (g)	Mass of blank HNO_3 (g)	Total cocktail mass (g)	f_{aq}(%)
1	4330d	UGAB (n = 5)	9.762(7)	0.046(11)	0.53(6)	0.1166(5)	10.45(5)	6.2(6)
		HiF (n = 4)[a]	9.725(7)	0.044(2)	0.6(5)	0.1156(12)	10.5(5)	7(4)
	4330c	UGAB (n = 5)	9.761(8)	0.042(5)	0.49(4)	0.115(7)	10.40(5)	5.8(4)
		HiF (n = 5)	9.722(4)	0.042(2)	0.50(4)	0.113(11)	10.38(5)	5.9(5)
2	4330d	UGAB (n = 6)	9.760(7)	0.030(7)	0.463(17)	0.0945(7)	10.348(19)	5.4(2)
		HiF (n = 6)	9.727(2)	0.030(9)	0.47(4)	0.0947(7)	10.32(4)	5.4(3)
	4330c	UGAB (n = 6)	9.758(5)	0.038(18)	0.45(6)	0.098(12)	10.34(8)	5.3(6)
		HiF (n = 6)	9.725(2)	0.036(18)	0.459(18)	0.0949(12)	10.315(15)	5.4(2)

[a] One source, PuDHiF2, is omitted from the data. Another source in this set, PuDHiF5, had double the amount of SRM solution added, leading to large uncertainties for SRM aliquot mass, total cocktail mass, and aqueous fraction (f_{aq}) for this data set.

2.3. Gamma spectrometry

Two ampoules were used for gamma-ray spectrometry spectroscopy for impurity analysis; one ampoule of SRM 4330d was transferred from a NIST-3 to NIST-1 ampoule (Collé, 2019), and another ampoule was measured in its original NIST-3 ampoule container.

Analysis for photon-emitting impurities was performed using a high-purity intrinsic germanium detector: X-detector (length, L = 36 mm; diameter, ϕ = 43 mm), which is an n-type coaxial detector with a 0.5 mm Be window. Details about the detector and measurement procedure have been described previously (Pibida et al., 2006, 2007).

2.4. Alpha spectrometry

2.4.1. Mixing of spikes

An overview of the radiochemistry and production of the solid sources for alpha spectrometry is shown in Fig. 2. Three sources were prepared from a single SRM 4330d ampoule for analysis by alpha spectrometry. Each source contained an aliquot of SRM 4330d weighing approximately 0.5 g measured by the pycnometer method (Sibbens and Altzitzoglou, 2007) into LS vials. To each aliquot approximately 0.7 g of ^{242}Pu SRM 4334i (certified massic activity: 26.77 Bq g^{-1} on January 1, 2010; 18.7 Bq per aliquot) was added and approximately 1.0 g of a diluted sample of ^{243}Am SRM 4332c (0.165 Bq g^{-1} on May 19th, 1995; 0.165 Bq per aliquot) was added as tracers for Pu and Am, respectively. These mixtures were diluted to 5 mL with deionized (DI) H_2O, bringing the HNO_3 concentration to approximately 0.75 mol L^{-1}.

In addition to the three spiked sources of SRM 4330d, a 1 g aliquot of SRM 4334i was spiked with 0.05 g of the diluted SRM 4332c and subjected to separations chemistry in order to analyze for any ^{243}Am impurity in SRM 4334i that may be biasing the Am analysis in SRM 4330d from the SRM 4334i spike. This source was treated identically to the three SRM 4330d spiked sources.

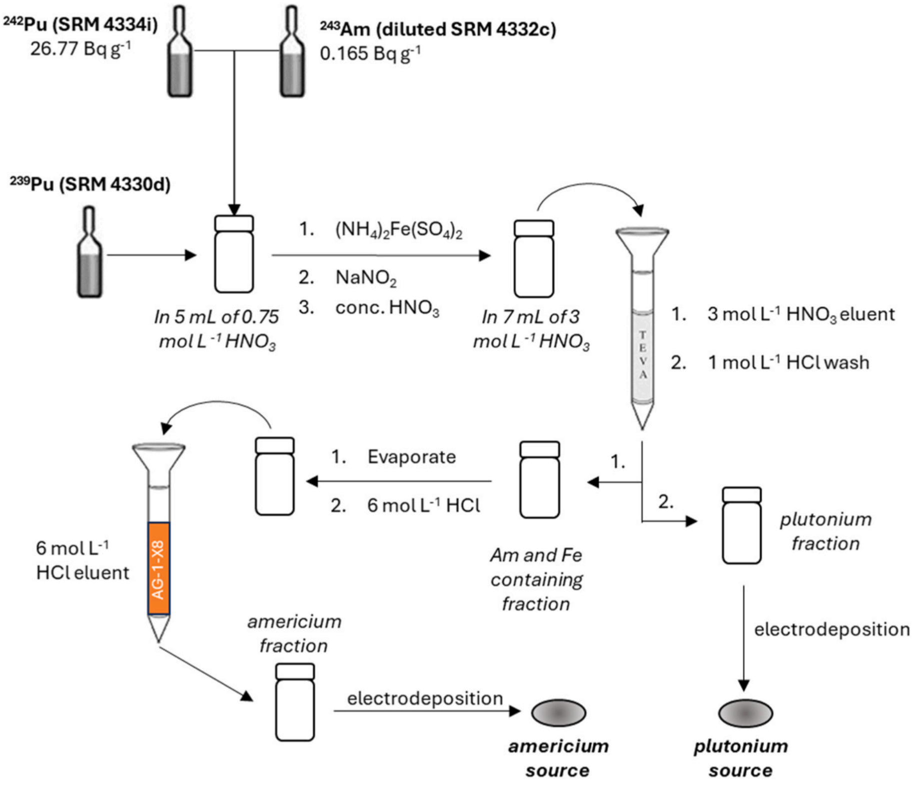

Fig. 2. Alpha spectrometry source preparation and radiochemical scheme.

2.4.2. Chemical equilibration and column extraction chromatography

To each source 50 mg $(NH_4)_2Fe(SO_4)_2$ (Mohr's salt) was added and allowed to dissolve for 5 min. This was followed by the addition 0.5 mL of 25 % $NaNO_2$ solution while swirling, which produced a dark green solution that faded to colorless over a minute. These steps ensure chemical equilibration of the analyte ^{239}Pu and the spike ^{242}Pu to the tetravalent state, which is necessary in the subsequent TEVA column separation step for adherence to the extractant.

The solutions were brought to a 3 mol L^{-1} HNO_3 concentration. Each source was subjected to separation of the Pu- and Am-containing fractions by loading the source onto a TEVA column (Eichrom Technologies, Lisle, IL, USA) in a 3 mol L^{-1} HNO_3 matrix, which is known to permit elution of the Am-containing fraction while retaining Pu (Horwitz et al., 1995). The purified Pu-containing fraction was collected separately from the Am-containing fraction by stripping the column with 1 mol L^{-1} HCl (15 mL).

2.4.3. Further purification of the americium fraction

The 3 mol L^{-1} HNO_3 eluate from the TEVA column was collected in 50 mL beakers and contains the ^{241}Am impurities of SRM 4330d, the ^{243}Am tracer, traces of organic TEVA extractant, and the Na^+ and Fe added as Mohr's salt and $NaNO_2$ for the chemical equilibration of the Pu, which must be removed prior to the electrodeposition step to avoid chemical interferences that would result in a loss in Am recovery. The solutions were treated with 5 mL of concentrated HCl and evaporated, resulting in some mild foaming as the oxidizing HCl/HNO_3 mixture reacts with NH_4^+ from the Mohr's salt, followed by precipitation of NaCl as the HCl concentration of the mixture increases during evaporation. Upon evaporating to approximately 1 mL of solution, the solutions were removed from the hot plate, brought to 20 mL with 1 mol L^{-1} HCl, and quantitatively transferred to 50 mL centrifuge tubes. To the solutions were added 2–3 mL of concentrated NH_4OH to precipitate $Fe(OH)_3$, which is known to carry actinides (Sill and Williams, 1981). The mixture was centrifuged, and the supernatant decanted to remove the excess Na^+. The solid pellet formed by centrifugation was dissolved in 9 mol L^{-1} HCl, and the resultant solution passed through an AG-1-X8 column, retaining the Fe as a $FeCl_4^-$ ion on the column. The anion exchange column eluate that contained the purified Am-fraction was evaporated to dryness and converted to nitrate form by addition of concentrated HNO_3 before proceeding to the electrodeposition step.

2.4.4. Solid source production and counting

Each sample was electrodeposited onto stainless steel disks using a modified procedure described previously (Glover et al., 1998). The samples were electrodeposited from solutions dissolved in a sodium hydrogen sulfate-sodium sulfate buffer ($NaHSO_4/Na_2SO_4$ ratio = 0.197; pH = 2.7).

Sources were counted on a Octête PC alpha spectrometer (Ametek Ortec, Oak Ridge, TN, USA) equipped with 8 identical 450 mm^2 ion-implanted Si detectors. Spectra were obtained using Ortec Maestro acquisition software (version 7.01). Electrodeposited sources (effective source diameter = 14 mm) were mounted 5 mm away from the detector for a counting efficiency of 24(1) %. Because the counting efficiency carries a large relative uncertainty (4.1 %), counting ratios against well-known, gravimetrically linked tracers were used to calculate the massic activities of species in the sample, circumventing the need to determine absolute activities of the alpha sources.

Chemical recoveries of plutonium and americium could be determined using the known counting efficiency for electrodeposited sources of 24(1) % and the known amount of activity of tracers ^{242}Pu and ^{243}Am added to the samples prior to extraction chromatography. In all cases, solid sources produced by the procedure had enough activity to reliably measure counting ratios. Recoveries varied from sample to sample and were dependent on the element. Plutonium recoveries were modest, ranging from 40 to 70 %. Americium recoveries were typically higher (>95 %–80 %) with the exception of one sample, where recoveries were <10 %.

2.5. Mass spectrometry

Two series of mass spectrometry measurements were made. The first series of measurements consisted of an isotope composition (IC) sample and two samples spiked with a high purity ^{244}Pu tracer (Essex et al., 2020) for isotope dilution mass spectrometry (IDMS). The second series was an IDMS comparison of three SRMs — the new SRM 4330d, the previous SRM 4330c, and ^{242}Pu SRM 4334i — consisting of two IDMS samples of each SRM. The second measurement series was conducted to obtain additional IDMS data on SRM 4330d as well as to validate the IDMS procedure using two well-characterized samples of different isotopes of plutonium.

In the first measurement series, one ampoule of SRM 4330d was opened and subdivided into 4 containers that were cleaned for ultra-low contamination chemistry: 2-perfluoroalkoxy (PFA) Teflon vials that had been pre-spiked with subsamples of the ^{244}Pu IDMS tracer and 2 high-density polyethylene (HDPE) bottles — one bottle for the IC sample and a second bottle for the remainder of the ampoule to be sampled from for the second mass spectrometry measurement series. The cleaning procedure for the labware included handwashing with Micro 90 laboratory soap, rinsing in 18.2 MΩ cm deionized water, soaking in 2 mol L^{-1} laboratory grade HNO_3, and rinsing again with 18.2 MΩ cm deionized water. Finally, the labware was conditioned with 2 rounds of fluxing with 8 mol L^{-1} Optima brand double-distilled HNO_3 by adding approximately 2 mL of acid to each container and setting it on a hotplate set to 90 °C for at least 24 h. All subsamples were prepared by mass using a low-density polyethylene (LDPE) pycnometer method (Sibbens and Altzitzoglou, 2007).

The IC sample was diluted to an approximate mass fraction of 0.5 ng g^{-1} of ^{239}Pu (based on conversion of approximately 38 Bq g^{-1} ^{239}Pu, which corresponds to 16.5 ng g^{-1}) with a mixed acid solution of 0.5 mol L^{-1} HNO_3 and 0.005 mol L^{-1} HF. To assure isotopic homogeneity of the mixed ^{239}Pu and ^{244}Pu solutions in the IDMS samples, each was treated to achieve the same valence state for plutonium. Approximately 3.7 mL of 8 mol L^{-1} HNO_3 was added to each vial to bring the solution to a molarity of 7 mol L^{-1} HNO_3. Then, 20 μL of 30 % H_2O_2 was added to each vial to reduce any higher-valent Pu(VI) or Pu(V) present to the Pu (IV) state (Marsh and Gallegos, 1987). The vials were capped and allowed to sit at room temperature overnight. After allowing the solution to equilibrate, the vials were opened and set on a hotplate to evaporate slowly (hot plate temperature <100 °C) until approximately 0.5 mL of solution remained. The solutions were then transferred to 60 mL HDPE bottles and diluted to an approximate ^{239}Pu mass fraction of 0.4 ng g^{-1} with the solution of 0.5 mol L^{-1} HNO_3 and 0.005 mol L^{-1} HF.

All samples in the second measurement series were treated identically to the two IDMS samples in the first measurement series, with a target final concentration of each major Pu isotope to be 0.5 ng g^{-1} in final solution.

Isotope-amount ratio analyses were performed on a ThermoFisher Element II mass spectrometer (ThermoFisher, Waltham, MA, USA) equipped with single secondary electron multiplier. The isotope composition for the ^{239}Pu IC and the IDMS samples were all analyzed using essentially the same measurement protocols. An equal atom ^{239}Pu – ^{242}Pu isotope CRM 128 (Crawford et al., 1990) was analyzed in sequence with the samples to correct for instrumental mass fractionation using the certified $n(^{242}\mathrm{Pu})/n(^{239}\mathrm{Pu})$ ratio and an exponential fractionation correction algorithm (Russell et al., 1978). All sample and standard analyses were bracketed by blank measurements of the 0.5 mol L^{-1} HNO_3 and 0.005 mol L^{-1} HF solution used to dilute the IC and IDMS samples. Due to the tendency of plutonium to adhere to the inlet system of the ICP-MS, an 8-min wash procedure using the same 0.5 mol L^{-1} HNO_3 and 0.005 mol L^{-1} HF solution was performed before each blank measurement. Instrument parameters for the analyses are provided in Table 2. Note that m/z = 235 and m/z = 238 were also measured on the

IC sample to estimate the amount of uranium and/or ^{238}Pu that was in the samples. It is important to consider contributions of ^{238}U^{1}H as a possible source isobaric interference when measuring ^{239}Pu, however in this case the count rates at $m/z = 238$ were sufficiently low to ignore this contribution (*vide infra*).

3. Results and discussion

3.1. Liquid scintillation

A summary of the LSC data is presented in Table 3. The results of the LS Comparison 1 found a massic counting rate — measured in counts per second per gram (s^{-1} g^{-1}) — for SRM 4330d to be 38.64(12) s^{-1} g^{-1}. A direct comparison with SRM 4330c with a certified massic activity (38.41(18) Bq g^{-1} ($k = 2$) at reference time of 1200 EST, May 1, 2009; 38.39 Bq g^{-1} at reference time for SRM 4330d of 1200 EST, July 28, 2024) was made to validate the LS counting method and potentially provide a means of correcting the LS counting data for SRM 4330d. However, SRM 4330c in LS Comparison 1 was found to have a massic counting rate of 37.99(13) s^{-1} g^{-1}, which differs by −1.0 % from the certified value for SRM 4330c and falls outside the expanded uncertainty but agree within 3-sigma. This was a statistically surprising result that warranted enough concern to prompt a second comparison experiment (LS Comparison 2).

There was essentially no difference between the measured massic counting rate of SRM 4330d between the two LS comparisons. The value of LS Comparison 2 for SRM 4330c was 38.38(13) s^{-1} g^{-1}, which differs from LS Comparison 1 by 1.0 % and agrees with the SRM certificate value within −0.02 %. However, because the isotopic composition of SRM 4330c differs greatly from SRM 4330d, a correction of the massic counting rate of SRM 4330d based on the comparison to SRM 4330c would not be an appropriate approach. Thus, a correction based on the impurity fractions obtained through secondary methods was sought.

3.2. Gamma spectrometry

Measurements of two ampoules of SRM 4330d only provided a small signal for ^{241}Am. The values for the two samples were disparate, but near the limit of detection (0.05 Bq s^{-1} g^{-1} for a 5 g sample of ^{241}Am measured at 59.5 keV) and thus carry substantial uncertainty: 0.15(2) Bq g^{-1} for the NIST-1 ampoule sample and 0.07(3) Bq g^{-1} for the NIST-3 ampoule sample.

3.3. Alpha spectrometry

The results of the Pu-fraction alpha sources are shown in Table 4. Alpha spectrometry is unable to resolve ^{239}Pu and ^{240}Pu and thus can only be measured as a combined ^{239,240}Pu activity ratio against the ^{242}Pu tracer. The combined activity $A_{239,240\mathrm{Pu}}$ in SRM 4330d was found to be 37.75(13) Bq g^{-1}. In addition, a small quantity of ^{238}Pu was observable and measured against the ^{242}Pu tracer to be 0.158(2) Bq g^{-1}. An internal isotopic ratio $A_{238\mathrm{Pu}}/A_{239,240\mathrm{Pu}}$ was also measured directly in all Pu-fraction samples, which was found to be 0.00419(5) and was used in the impurity correction to the LS data. A characteristic alpha spectrum from a purified Pu-fraction source is shown in Fig. 3.

Table 2
Element II ICP-MS instrument parameters for ^{239}Pu measurements.

Scan Order	1	2	3	4	5	6
IC	^{235}U	^{238}U	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
IDMS	^{239}Pu	^{238}Pu ^{240}Pu	^{244}Pu		^{241}Am	
Sample and Skimmer Cone			Ni	Instrument Analysis Gas		Ar
Passes			5	- Coolant Gas (L min^{-1})		16.0
Runs			5	- Sample Gas (L min^{-1})		0.95
Integration Time (s)			0.20	- Auxiliary Gas (L min^{-1})		0.66
Nebulizer			PFA	High Voltage (V)		−7932
- Sample Uptake (μL min^{-1})			100	RF Power (W)		1250

Table 3
Summary of LS Comparison experiment results.

LS Comparison	SRM 4330c (s^{-1} g^{-1})	SRM 4330d (s^{-1} g^{-1})
1	37.99(13)	38.63(12)
2	38.38(13)	38.64(12)
% difference[a]	+1.0	0.0

[a] difference is calculated as change from LS Comparison 1 to LS Comparison 2.

Table 4
Alpha spectrometry results for plutonium fraction measured against ^{242}Pu tracer.

Sample ID	$A(^{239,240}\mathrm{Pu})$ (Bq g^{-1})	$A(^{238}\mathrm{Pu})$ (Bq g^{-1})	$A(^{238}\mathrm{Pu})/A(^{239,240}\mathrm{Pu})$
AS-Pu-1	37.73(14)	0.157(3)	0.00417(7)
AS-Pu-2	37.73(12)	0.1572(13)	0.00417(3)
AS-Pu-3	37.78(12)	0.1592(13)	0.00422(3)
Average	37.75(13)	0.158(2)	0.00419(5)

Alpha spectrometry on the Am-fractions revealed a small quantity of ^{241}Am present in SRM 4330d, with an average A = 0.069 Bq g^{-1} determined *on the date of measurement* (March 9, 2024 for Sample AS-Am-1, 9/18/2024 for both Sample AS-Am-2 and AS-Am-3). A simple, unsupported decay correction to the SRM reference time (7/28/2024 12:00 EST) would have a negligible effect on the value for $A_{241\mathrm{Am}}$ ($T_{1/2}$ = 432.6 a; difference in the measurement time from reference time = 37–52 days). However, the presence of parent ^{241}Pu complicates the decay correction and is discussed in greater detail (*vide infra*).

3.4. Mass spectrometry

The results for the $n(^{240}\mathrm{Pu})/n(^{239}\mathrm{Pu})$ measurements of SRM 4330d are shown in Table 5. In total, 5 samples of SRM 4330d were measured by mass spectrometry, with one sample measured as dilution of SRM 4330d and the other 4 as IDMS samples that were diluted and spiked with a ^{244}Pu tracer of precisely known composition (Essex et al., 2020). All mass spectrometry measurements were corrected for background, mass bias, and — in the case of IDMS samples — spike corrected for known isotopic ratios $n(^{239}\mathrm{Pu})/n(^{244}\mathrm{Pu})$ and $n(^{240}\mathrm{Pu})/n(^{244}\mathrm{Pu})$ of the tracer.

The mean $n(^{240}\mathrm{Pu})/n(^{239}\mathrm{Pu})$ ratio across the 5 samples was measured as 0.00848(6), with the uncertainty determined from the root mean square of the internal and external standard deviations of the sample measurements. From the atom ratio $n(^{240}\mathrm{Pu})/n(^{239}\mathrm{Pu})$ measured by mass spectrometry it is possible to convert to an activity ratio $A_{240\mathrm{Pu}}/A_{239\mathrm{Pu}}$ using DDEP-accepted half-life data for ^{239}Pu ($T_{1/2} = 24100(11)$ a; $\lambda_{239\mathrm{Pu}} = 2.8761(13) \times 10^{-5}$ a^{-1}) and ^{240}Pu ($T_{1/2} = 6561(7)$ a; $\lambda_{240\mathrm{Pu}} = 1.0565(11) \times 10^{-4}$ a^{-1}), which was determined to be 0.0311(2). This value is in good agreement with the activity ratio provided in the stock material certificate of calibration from EZA.

The IC measurements revealed a $n(^{241}\mathrm{Pu}+^{241}\mathrm{Am})/n(^{239}\mathrm{Pu})$ ratio of $7.48(6) \times 10^{-5}$, which is larger than the anticipated atom ratio derived from the activity of ^{241}Pu and ^{241}Am provided from the stock material certificate of calibration. This ratio can be used in conjunction with the activity of ^{241}Am determined by alpha spectrometry to approximate the amount of ^{241}Pu present (*vide infra*).

IC measurements were not able to detect a measurable amount of ^{242}Pu. Additionally, a $n(^{238}\mathrm{U}+^{238}\mathrm{Pu})/n(^{239}\mathrm{Pu})$ ratio could be measured, and was determined to be 0.00047(4). Accounting for the $n(^{238}\mathrm{Pu})/n(^{239}\mathrm{Pu})$ ratio inferred from alpha spectrometry, the contribution of ^{238}Pu to $n(^{238}\mathrm{U}+^{238}\mathrm{Pu})/n(^{239}\mathrm{Pu})$ is negligible. However, assuming $n(^{238}\mathrm{U}+^{238}\mathrm{Pu})/n(^{239}\mathrm{Pu})$ approximates to $n(^{238}\mathrm{U})/n(^{239}\mathrm{Pu})$, an exceedingly small activity ratio of $A_{238\mathrm{U}}/A_{239\mathrm{Pu}}$ can be calculated to be on the

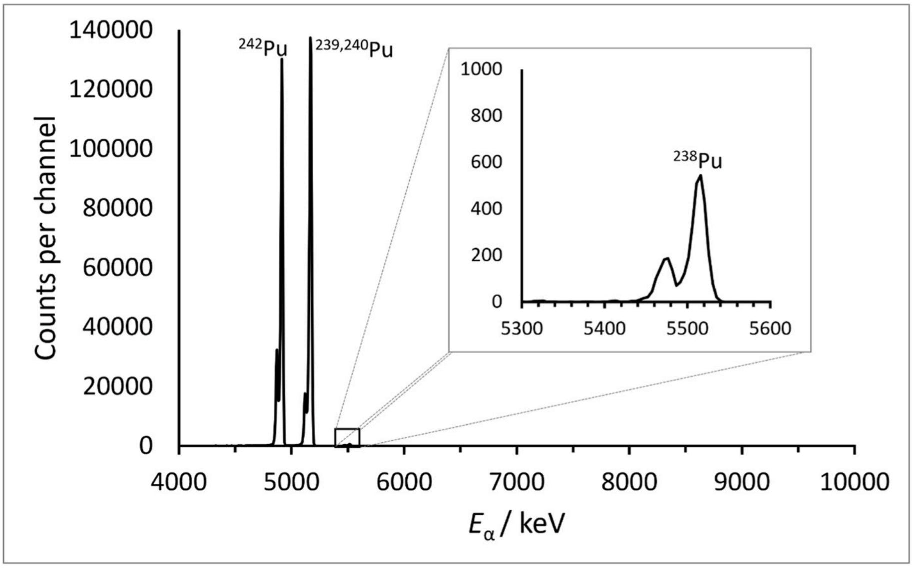

Fig. 3. Alpha spectrum for AS-Pu-1. Inset shows zoom-in of ^{238}Pu peak.

Table 5
Isotopic composition determinations for SRM 4330d by mass spectrometry.

Sample ID	N_m	$n(^{240}Pu)/n(^{239}Pu)$	$n(^{241}Pu+^{241}Am)/n(^{239}Pu)$
IC	6	0.00841(5)	$7.48(6) \times 10^{-5}$
IDMS-d1	2	0.00846(7)	Not measured
IDMS-d2	2	0.00849(4)	Not measured
IDMS-d3	3	0.00846(3)	Not measured
IDMS-d4	3	0.00855(10)	Not measured
	Average =	**0.00848(6)**	$\mathbf{7.48(6) \times 10^{-5}}$

Table 6
IDMS results against ^{244}Pu tracer.

SRM	Sample ID	N_m [a]	Activity (Bq g^{-1}) measured per sample	Average Activity (Bq g^{-1}) per SRM	Activity expected (Bq g^{-1})[b]	% difference from expected value
4330d	IDMS-d1	2	37.07(16)	37.6(5)	36.6	2.8
	IDMS-d2	2	38.10(13)			
	IDMS-d3	3	37.94(11)			
	IDMS-d4	3	37.46(9)			
4330c	IDMS-c1	3	39.3(2)	39.8(8)	38.39(13)	3.7
	IDMS-c2	3	40.3(3)			
4334i	IDMS-i1	4	27.71(19)	27.9(3)	26.77(9)	4.1
	IDMS-i2	4	28.0(3)			

[a] N_m = number of measurements per sample.

[b] Expected activity for 4330d is based on the final correction of the LS data (*vide infra*); expected activities for 4330c and 4334i are certificate values.

order of nBq g^{-1} and can be safely neglected.

The IDMS samples were used as confirmatory measurements on the ^{239}Pu massic activity in SRM 4330d measured by LS and alpha spectrometry. In addition, control measurements of the previously issued ^{239}Pu SRM 4330c and a^{242}Pu SRM 4330i were measured against the ^{244}Pu spike in order to validate the IDMS procedure. The results of the IDMS experiments are summarized in Table 6. Across all measurements, the IDMS procedure overestimates the massic activity of the nuclide of interest in every sample between 2 and 45 %. Furthermore, there is a relatively large standard deviation between the samples relative to the deviation measured within each sample. This is indicative of a possible flaw in the chemical equilibration procedure used: if there is a variable loss of ^{244}Pu in every sample, then one would expect to observe a consistent overestimation in the value of the measurand accompanied with a large standard deviation in the measured value across samples relative to the internal error for each individual sample. Further work is ongoing to determine the nature of the error in the IDMS measurement and to improve upon the IDMS procedure for future confirmatory measurements by mass spectrometry.

4. Discussion and analysis

4.1. Combining the alpha spectrometry and mass spectrometry results

A more useful quantity for the alpha spectrometry corrections is the activity of either ^{239}Pu or ^{240}Pu relative to the combined ^{239,240}Pu peak unresolvable in the alpha spectrum, which can be obtained simply by eq. (1):

$$\frac{A_{239Pu}}{A_{239Pu}+A_{240Pu}}=1-\frac{A_{240Pu}}{A_{239Pu}+A_{240Pu}} \qquad \text{(eq. 1)}$$

where $A_{240\mathrm{Pu}}/A_{239,240\mathrm{Pu}}$ and $A_{239\mathrm{Pu}}/A_{239,240\mathrm{Pu}}$ are found to be 3.0 % and 97.0 %, respectively. This correction can be applied to the measured $A_{238\mathrm{Pu}}/A_{239,240\mathrm{Pu}}$ value to find $A_{238\mathrm{Pu}}/A_{239\mathrm{Pu}} = 0.00432(6)$. This value will be used in the impurity correction to the 4πα LS value (*vide infra*).

4.1.1. ^{241}Am and ^{241}Pu

The activity for ^{241}Am (and by extension ^{241}Pu) required isotope dilution measurements by alpha spectrometry after radiochemical separation, and thus a direct ratio to ^{239}Pu cannot be measured for any individual mass 241 isobars. However, the sum of these isobars is known via mass spectrometry and thus provides a means of calculating an estimate for the value of $A_{241\mathrm{Pu}}$ at the measurement time by eqs. (2) and (3):

$$A_{241Pu}=\frac{\lambda_{241Pu}A_{239Pu}}{\lambda_{239Pu}}\times\frac{n(^{241}Pu)}{n(^{239}Pu)} \qquad \text{(eq. 2)}$$

$$\frac{n(^{241}Pu)}{n(^{239}Pu)} = \frac{n(^{241}Am) + n(^{241}Pu)}{n(^{239}Pu)} - \frac{A_{241Am}\lambda_{239Pu}}{A_{239Pu}\lambda_{241Am}} \qquad \text{(eq. 3)}$$

It is noted that the time-dependance on the ratio $n(^{241}\text{Am}+^{241}\text{Pu})/n(^{239}\text{Pu})$ is dependent on the relative amounts of ^{241}Am and ^{241}Pu. In order to account for the uncertainty in how the ratio may change from the date of mass ratio measured (July 17th, 2024) and the date of the ^{241}Am activity measurement (September 3rd-18th, 2024), a Type B uncertainty component was added to account for the difference in this ratio expected when decayed to the ^{241}Am activity measurement date if ^{241}u was composed of either 1) all ^{241}Am or 2) all ^{241}Pu. Because the change was negligible in the time interval that all measurements were made, the value of $n(^{241}\text{Am}+^{241}\text{Pu})/n(^{239}\text{Pu})$ was kept constant, and the Type B expanded relative uncertainty was determined to be <0.1 % in all cases. $n(^{241}\text{Pu})/n(^{239}\text{Pu})$ was found to be $4.09(16) \times 10^{-5}$, and upon converting to a massic activity $A_{241\text{Pu}}$ using the value $A_{239\text{Pu}}$ found from isotope dilution alpha spectrometry and accepted DDEP half-life data, a value for $A_{241\text{Pu}}$ of 2.53(12) Bq g^{-1} was found after decay correction.

Decay-corrected massic activity calculations for the individual ^{241}Am samples, A^0_{241Am}, were made after approximation of the parent ^{241}Pu activity in each sample by rearrangement of the two-component Bateman equation, eq. (4):

$$A^0_{241Am} = \frac{A^t_{241Am} - \frac{A^0_{241Pu}\lambda_{241Am}}{\lambda_{241Am}-\lambda_{241Pu}}\left(e^{-\lambda_{241Pu}t} - e^{-\lambda_{241Am}t}\right)}{e^{-\lambda_{241Am}t}} \qquad \text{(eq. 4)}$$

where A^t_{241Am} is the activity of ^{241}Am found by alpha spectrometry against the ^{243}Am tracer at the measurement time of the sample, A^0_{241Pu} is the decay-corrected ^{241}Pu massic activity for the sample as described previously, and t is the time between the ^{241}Am alpha spectrometry measurement and the reference time. Table 7 shows the values for the individual decay corrections for ^{241}Pu and ^{241}Am, as well as the resultant mean A^0_{241Am}, which was found to be 0.069(4) Bq g^{-1}.

The counting efficiency for ^{241}Pu by LS counting was not determined through CIEMAT/NIST efficiency tracing (CNET) for this experiment, but can be estimated using the following assumptions: 1) the composition of SRM 4330d consists only of ^{239}Pu and alpha-emitting impurities ^{238}Pu, ^{240}Pu, and ^{241}Am, with ^{241}Pu being the only beta-emitting impurity, and 2) the activities of the alpha-emitting impurities can be reasonably approximated through secondary standardization methods. Using the activities $A_{239,240\text{Pu}}$, $A_{238\text{Pu}}$, and $A_{241\text{Am}}$ determined by alpha spectrometry and mass spectrometry and subtracting these values from the LS counting rate R_{LS}, a remaining count rate attributed to ^{241}Pu ($R_{241\text{Pu}}$) can be calculated by eq. 5, and a^{241}Pu LS counting efficiency ($\varepsilon_{241\text{Pu}}$) by eq. (6):

$$R_{241Pu} = R_{LS} - \left(A_{239,240Pu} + A_{238Pu} + A_{241Am}\right) \qquad \text{(eq. 5)}$$

$$R_{241Pu} = A_{241Pu}\varepsilon_{241Pu} \qquad \text{(eq. 6)}$$

From the above equations, the remaining count rate $R_{241\text{Pu}}$ is found to be 0.7(3) s^{-1} g^{-1}, and $\varepsilon_{241\text{Pu}}$ approximated to 26(11) %. This value is consistent with efficiencies computed from CNET data in a previous standardization of a^{241}Pu SRM 4340b of similar LS cocktail composition and measured external standard quench parameter. The large uncertainties associated with the counting rate and ^{241}Pu efficiency reflect the large propagation of error in the chain of calculations required for the approximation.

Table 7
Summary of decay correction of ^{241}Am accounting for ingrowth due to ^{241}Pu.

Sample	Activity vs ^{243}Am tracer (Bq g^{-1})	Measurement reference time, t (d)	A^t_{241Pu} (Bq g^{-1})	A^o_{241Pu} (Bq g^{-1})	A^0_{241Am} (Bq g^{-1})
Am-1	0.0705(10)	37.0	2.47(6)	2.49(6)	0.0701(10)
Am-2	0.0654(11)	52.0	2.63(6)	2.65(6)	0.0648(11)
Am-3	0.072(5)	52.0	2.44(15)	2.46(15)	0.071(5)
			Average =	**2.53(12)**	**0.069(4)**

4.2. *Impurity correction to the LS data*

The primary standardization by LS provides a total massic count rate for the SRM (R_{LS}) that needs to be corrected using known impurity ratios relative to the nuclide of interest ^{239}Pu. The impurity correction is described by eq. (7) and eq. (8):

$$A_{239Pu} = \frac{R_{LS}}{C_i} \qquad \text{(eq. 7)}$$

$$C_i = 1 + \frac{A_{240Pu}}{A_{239Pu}} + \frac{A_{238Pu}}{A_{239Pu}} + \frac{A_{241Am}}{A_{239Pu}} + \frac{R_{241Pu}}{A_{239Pu}} \qquad \text{(eq. 8)}$$

where C_{i} is the impurity correction factor. Using eq. (7), the final value for ^{239}Pu activity was determined to be 36.6(6) Bq g^{-1} ($k = 2$), which corresponds to a 1.6 % expanded relative uncertainty. This is significantly higher than typical standard reference solutions and illustrates the challenge of correcting 4πα LSC data by secondary methods when the impurities are in large quantity. Table 8 shows the uncertainty budget for the activity of ^{239}Pu standardized by 4πα LS, and Table 9 shows a detailed breakdown of the uncertainty in C_{i}.

5. Summary and conclusions

A^{239}Pu solution containing 36.6(6) Bq g^{-1} ($k = 2$) of ^{239}Pu as well as > 5 % impurity by activity was standardized by 4πα liquid scintillation relying heavily on mass and alpha spectrometry measurements for impurity correction. The complementary nature of the alpha and mass spectrometry allowed for reliable impurity assessment of alpha-emitting plutonium isotopes ^{238}Pu and ^{240}Pu. While the determination of the activity of beta-emitting ^{241}Pu through indirect methods provided a large uncertainty, this work allowed the activity to be accounted for. Radiochemical separation of americium and plutonium prior to analysis by mass spectrometry should lead to improvement of the ^{241}Pu determination, and this is being explored further.

CRediT authorship contribution statement

A.J. Pearce: Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Data curation. **M.R. Bodine:** Writing – review & editing, Investigation, Formal analysis. **R. Collé:** Validation, Supervision, Methodology, Investigation, Formal analysis, Conceptualization. **R. Essex:** Writing – review & editing, Writing – original draft, Methodology, Investigation. **J. La Rosa:** Supervision, Methodology, Investigation. **L. Laureano-Pérez:** Validation, Supervision, Software, Resources, Project administration, Methodology, Investigation, Funding acquisition, Conceptualization. **L. Pibida:** Software, Methodology, Investigation, Formal analysis. **M.A. Tyra:** Writing – review & editing, Investigation.

Disclaimer

Certain commercial equipment, instruments, or materials are identified in this paper to foster understanding. Such identification does not imply recommendation by the National Institute of Standards and Technology, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose.

Table 8

Uncertainty budget for the value of the massic activity of ^{239}Pu ($A_{^{239}Pu}$) in SRM 4330d. Assessments were of either type A (evaluation by statistical methods) or type B (evaluation by other methods)(Taylor and Kuyatt, 1994).

Uncertainty component		Assessment Type	Relative standard uncertainty contribution on massic activity of ^{239}Pu (%)
1	LS measurement precision: standard deviation for n = 5 mean determinations as obtained with 2 different cocktail compositions (4–6 sources of each composition) measured 2 times in 2 different LS counters (105 determinations in all).	A	0.3
2	Background LS measurement variability; wholly embodied in component 1	A	–
3	Impurity correction factor (C_i) uncertainty (see Table 9 for breakdown)	B	0.7
4	Gravimetric determinations for LS sources; estimated from calibration data and tests	B	0.02
5	Decay correction due to the decay time interval and 0.046 % uncertainty in half-life.	B	1 x 10^{-8}
6	Live time determinations for LS counting time intervals, includes uncorrected dead time effects; assumed from specified tolerance limits of counters' gated oscillators	B	0.07
7	LS detection efficiency, including wall effects loss and extrapolation to zero energy; estimated from nuclear data and tests.	B	0.07
8	Alpha decay probability; assumed from nuclear decay data based on % of spontaneous fission	B	3 x 10^{-10}
9	LS non-detection of 26 min ^{235m}U; estimated from nuclear decay data and tests of LS counter detection threshold	B	<0.001
10	Correction for ingrowth of ^{235}U; estimated from nuclear data	B	3 x 10^{-8}
Relative combined standard uncertainty			**0.77**
Relative expanded uncertainty (k=2)			**1.54**

Table 9

Uncertainty budget for impurity correction factor (C_i).

Term		Value	u	%u
1	A_{240Pu}/A_{239Pu},	0.0311	0.0002	0.6
2	A_{238Pu}/A_{239Pu}	0.00432	0.00006	1.3
3	A_{241Am}/A_{239Pu}	0.00187	0.00010	5.5
4	R_{241Pu}/A_{239Pu}	0.018	0.008	42.6
$\boldsymbol{C_i}$		**1.056**	**0.008**	**0.7**

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

Jacqueline Mann provided valuable insight into metrological considerations for mass spectrometry. Special thanks for helpful words of advice and encouragement to Denis Bergeron, Max Carlson, Ryan Fitzgerald, Lisa Karam, Svetlana Nour, and Brian Zimmerman.

Data availability

Data will be made available on request.

References

Bièvre, P.D., Verbruggen, A., 1999. A new measurement of the half-life of ^{241}Pu using isotope mass spectrometry. Metrologia 36 (1), 25. https://doi.org/10.1088/0026-1394/36/1/5.

Braysher, E., Russell, B., Read, D., 2020. Application of plasma mass spectrometry for half-life measurement of medium and long-lived radionuclides. J. Phys. Conf. 1643 (1), 012207. https://doi.org/10.1088/1742-6596/1643/1/012207.

Cassette, P., Chartier, F., Isnard, H., Fréchou, C., Laszak, I., Degros, J.P., Bé, M.M., Lépy, M.C., Tartes, I., 2010. Determination of ^{93}Zr decay scheme and half-life. Appl. Radiat. Isot. 68 (1), 122–130. https://doi.org/10.1016/j.apradiso.2009.08.011.

Collé, R., 2019. Ampoules for Radioactivity Standard Reference Materials. NISTIR 8254).

Crawford, D.W., Gradle, C.G., Soriano, M.D., 1990. The Production and Certification of a Plutonium equal-atom Reference Material: NBL CRM 128. NBL-316).

Crozet, M., Roudil, D., Maillard, C., Rivier, C., Jakopič, R., Fankhauser, A., Aregbe, Y., Richter, S., 2019. Contribution of an interlaboratory comparison to the certification of the STAM/IRMM-0243 ^{243}Am reference material. J. Radioanal. Nucl. Chem. 319 (3), 717–725. https://doi.org/10.1007/s10967-018-06404-6.

Dersch, R., 2004. Primary and secondary measurements of ^{222}Rn. Appl. Radiat. Isot. 60 (2), 387–390. https://doi.org/10.1016/j.apradiso.2003.11.046.

Essex, R.M., Williams, R.W., Treinen, K.C., Hubert, A., Humphrey, M.A., Inglis, J.D., Kinman, W.S., Maassen, J., Penkin, M.V., Steiner, R.E., 2020. A highly-enriched ^{244}Pu reference material for nuclear safeguards and nuclear forensics measurements. J. Radioanal. Nucl. Chem. 324 (1). https://doi.org/10.1007/s10967-020-07075-y.

Glover, S.E., Filby, R.H., Clark, S.B., Grytdal, S.P., 1998. Optimization and characterization of a sulfate based electrodeposition method for alpha-spectroscopy of actinide elements using chemometric analysis. J. Radioanal. Nucl. Chem. 234 (1–2), 213–218. https://doi.org/10.1007/BF02389774.

Gray, L.W., Holliday, K.S., Murray, A., Thompson, M., Thorp, D.T., Yarbro, S., Venetz, T. J., 2015. Separation of Plutonium from Irradiated Fuels and Targets. LLNL-TR-677668).

Horwitz, E.P., Dietz, M.L., Chiarizia, R., Diamond, H., Maxwell, S.L., Nelson, M.R., 1995. Separation and preconcentration of actinides by extraction chromatography using a supported liquid anion exchanger: application to the characterization of high-level nuclear waste solutions. Anal. Chim. Acta 310 (1), 63–78. https://doi.org/10.1016/0003-2670(95)00144-O.

Kossert, K., Jörg, G., Gostomski, C.L.v., 2013. Experimental half-life determination of ^{176}Lu. Appl. Radiat. Isot. 81, 140–145. https://doi.org/10.1016/j.apradiso.2013.03.033.

Laureano-Pérez, L., Collé, R., Fitzgerald, R., Outola, I., Pibida, L., 2007. A liquid-scintillation-based primary standardization of ^{210}Pb. Appl. Radiat. Isot. 65 (12), 1368–1380. https://doi.org/10.1016/j.apradiso.2007.06.012.

Marouli, M., Pommé, S., Jobbágy, V., Stroh, H., Van Ammel, R., Fankhauser, A., Jakopič, R., Richter, S., Aregbe, Y., Crozet, M., Maillard, C., Rivier, C., Roudil, D., 2020. Absolute and relative measurement of the ^{243}Am half-life. J. Radioanal. Nucl. Chem. 326 (3), 1785–1793. https://doi.org/10.1007/s10967-020-07450-9.

Marsh, S.F., Gallegos, T.D., 1987. *Chemical Treatment of Plutonium with Hydrogen Peroxide Before Nitrate Anion Exchange Processing* (LA-10907).

Moody, K.J., Grant, P.M., Hutcheon, I.D., 2014. Nuclear Forensic Analysis, second ed. CRC Press. https://doi.org/10.1201/b17863.

Nedjadi, Y., Bailat, C., Caffari, Y., Froidevaux, P., Wastiel, C., Kivel, N., Guenther-Leopold, I., Triscone, G., Jaquenod, F., Bochud, F., 2012. A new measurement of the half-life of ^{166m}Ho. Appl. Radiat. Isot. 70 (9), 1990–1996. https://doi.org/10.1016/j.apradiso.2012.02.063.

Parsons-Davis, T., Wimpenny, J., Keller, C.B., Thomas, K., Samperton, K.M., Renne, P.R., Mundil, R., Moody, K., Knight, K., Kristo, M.J., Williams, R., 2018. New measurement of the ^{238}U decay constant with inductively coupled plasma mass spectrometry. J. Radioanal. Nucl. Chem. 318 (1), 711–721. https://doi.org/10.1007/s10967-018-6148-y.

Pibida, L., Hsieh, E., Fuentes-Figueroa, A., Hammond, M.M., Karam, L., 2006. Software studies for germanium detectors data analysis. Appl. Radiat. Isot. 64 (10–11), 1313–1318. https://doi.org/10.1016/j.apradiso.2006.02.076.

Pibida, L., Nafee, S.S., Unterweger, M., Hammond, M.M., Karam, L., Abbas, M.I., 2007. Calibration of HPGe gamma-ray detectors for measurement of radioactive noble gas sources. Appl. Radiat. Isot. 65 (2), 225–233. https://doi.org/10.1016/j.apradiso.2006.07.002.

Pommé, S., 2007. Methods for primary standardization of activity. Metrologia 44 (4), S17. https://doi.org/10.1088/0026-1394/44/4/S03.

Pommé, S., 2022. Radionuclide metrology: confidence in radioactivity measurements. J. Radioanal. Nucl. Chem. 331 (12), 4771–4798. https://doi.org/10.1007/s10967-022-08494-9.

Russell, W.A., Papanastassiou, D.A., Tombrello, T.A., 1978. Ca isotope fractionation on the Earth and other solar system materials. Geochem. Cosmochim. Acta 42 (8), 1075–1090. https://doi.org/10.1016/0016-7037(78)90105-9.

Sibbens, G., Altzitzoglou, T., 2007. Preparation of radioactive sources for radionuclide metrology. Metrologia 44 (4), S71–S78. https://doi.org/10.1088/0026-1394/44/4/S09.

Sill, C.W., Williams, R.L., 1981. Preparation of actinides for alpha spectrometry without electrodeposition. Anal. Chem. 53 (3), 412–415. https://doi.org/10.1021/ac00226a008.

Taylor, B.N., Kuyatt, C.E., 1994. Guidelines for Evaluating and Expressing the Uncertainty of NIST Measurement Results. NIST Technical Note 1297).

Varga, Z., Nicholl, A., Wallenius, M., Mayer, K., 2016. Remeasurement of ^{234}U half-life. Anal. Chem. 88 (5), 2763–2769. https://doi.org/10.1021/acs.analchem.5b04370.

Yang, J.L., Zhang, S.D., Ding, Y.Q., Shu, F.J., Zhang, J.L., 2010. A new value of ^{93}Zr half-life. Radiochim. Acta 98 (2), 59–63. https://doi.org/10.1524/ract.2010.1678(rca-RadiochimicaActa.