



National Institute of Standards & Technology Certificate

Standard Reference Material 4330B Plutonium-239 Radioactivity Standard

This Standard Reference Material (SRM) consists of radioactive plutonium-239 nitrate and nitric acid dissolved in 5 mL of distilled water. The solution is contained in a flame-sealed NIST borosilicate-glass ampoule. The SRM is intended for the calibration of alpha-particle counting instruments and for the monitoring of radiochemical procedures.

Radiological Hazard: The SRM ampoule contains plutonium-239 with a total activity of approximately 220 Bq. Plutonium-239 decays by alpha-particle emission. None of the alpha particles escape from the SRM ampoule. During the decay process X-rays and gamma rays with energies from 10 keV to 1 MeV are also emitted. Most of these photons escape from the SRM ampoule but their intensities are so small that they do not represent a radiation hazard. Approximate unshielded dose rates at several distances (as of the reference time) are given in note [a]*. The SRM should be used only by persons qualified to handle radioactive material.

Chemical Hazard: The SRM ampoule contains nitric acid (HNO_3) with a concentration of 3 moles per liter of water. The solution is corrosive and represents a health hazard if it comes in contact with eyes or skin. If the ampoule is to be opened to transfer the solution, the recommended procedure is given on page 2. The ampoule should be opened only by persons qualified to handle both radioactive material and strong acid solution.

Storage and Handling: The SRM should be stored and used at a temperature between 5 and 65 °C. The solution in an unopened ampoule should remain stable and homogeneous until at least December 2009.

The ampoule (or any subsequent container) should always be clearly marked as containing radioactive material. If the ampoule is transported it should be packed, marked, labeled, and shipped in accordance with the applicable national, international, and carrier regulations. The solution in the ampoule is a dangerous good (hazardous material) because of the radioactivity and because of the strong acid.

Preparation: This Standard Reference Material was prepared in the Physics Laboratory, Ionizing Radiation Division, Radioactivity Group, L.R. Karam, Group Leader. The overall technical direction and physical measurements leading to certification were provided by L.L. Lucas of the Radioactivity Group.

The support aspects involved in the preparation, certification, and issuance of this SRM were coordinated through the Standard Reference Materials Program by J.W.L. Thomas.

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March 2000

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Recommended Procedure for Opening the SRM Ampoule

- 1) If the SRM solution is to be diluted, it is recommended that the diluting solution have a composition comparable to that of the SRM solution.
- 2) Wear eye protection, gloves, and protective clothing and work over a tray with absorbent paper in it. Work in a fume hood. In addition to the radioactive material, the solution contains strong acid and is corrosive.
- 3) Shake the ampoule to wet all of the inside surface of the ampoule. Return the ampoule to the upright position.
- 4) Check that all of the liquid has drained out of the neck of the ampoule. If necessary, gently tap the neck to speed the process.
- 5) Holding the ampoule upright, score the narrowest part of the neck with a scribe or diamond pencil.
- 6) Lightly wet the scored line. This reduces the crack propagation velocity and makes for a cleaner break.
- 7) Hold the ampoule upright with a paper towel, a wiper, or a support jig. Position the scored line away from you. Using a paper towel or wiper to avoid contamination, snap off the top of the ampoule by pressing the narrowest part of the neck away from you while pulling the tip of the ampoule towards you.
- 8) Transfer the solution from the ampoule using a pycnometer or a pipet with dispenser handle. NEVER PIPETTE BY MOUTH.
- 9) Seal any unused SRM solution in a flame-sealed glass ampoule, if possible, to minimize the evaporation loss.

See also reference [4]*.

PROPERTIES OF SRM 4330B

Certified values

Radionuclide	Plutonium-239
Reference time	1200 EST, 15 November 1999
Massic activity of the solution [b]*	39.24 Bq·g⁻¹
Relative expanded uncertainty (k=2)	0.68 % [c] [d]
Solution density	(1.101 ± 0.002) g·mL⁻¹ at 21.4 °C [e]

Uncertified values

Physical Properties:			
Source description	Liquid in flame-sealed NIST borosilicate-glass ampoule		
Ampoule specifications	Body outside diameter	(16.5 ± 0.5) mm	
	Wall thickness	(0.60 ± 0.04) mm	
	Barium content	Less than 2.5%	
	Lead-oxide content	Less than 0.02%	
	Other heavy elements	Trace quantities	
Solution mass	Approximately 5.5 grams		
Chemical Properties:			
Solution composition	Chemical Formula	Concentration (mol·L ⁻¹)	Mass Fraction (g·g ⁻¹)
	H ₂ O	50	0.82
	HNO ₃	3.1	0.18
	HCl	0.02	0.0007
	²³⁹ Pu(NO ₃) ₄	8 × 10 ⁻⁸	5 × 10 ⁻⁸
Radiological Properties:			
Alpha-particle-emitting impurities	None detected [f]		
Photon-emitting impurities	None detected [g]		
Half lives used	Plutonium-239: (24110 ± 30) a [h] [5]		
Calibration method and measuring instrument(s)	Two 4π α liquid-scintillation counting systems		

EVALUATION OF THE UNCERTAINTY OF THE MASSIC ACTIVITY [c] [d]*

Input Quantity x , the source of uncertainty (and individual uncertainty components where appropriate)	Method Used To Evaluate $u(x)$, the standard uncertainty of x (A) denotes evaluation by statistical methods (B) denotes evaluation by other methods	Relative Uncertainty Of Input Quantity, $u(x)/x$, (%) [i]	Relative Sensitivity Factor, $ \partial y/\partial x_i \cdot$ (x/y) [j]	Relative Uncertainty Of Output Quantity, $u(y)/y$, (%) [k]
Massic alpha-particle emission rate, corrected for background and decay	Standard deviation of the mean for 18 sets of $4\pi\alpha$ liquid- scintillation measurements (A)	0.02	1.0	0.02
Half life of plutonium-239	Standard uncertainty of the half life (A)	0.11 [m]	0.0005 [n]	0.00005
Decay-scheme data	Standard uncertainty of the probability of decay by alpha- particle emission (A)	0.001	1.0	0.001
Gravimetric measurements	Estimated (B)	0.10	1.0	0.10
Live-time [p]	Estimated (B)	0.10	1.0	0.10
Alpha-particle detection efficiency of the liquid-scintillation counting systems [q]	Estimated (B)	0.27	1.0	0.27
Alpha-particle-emitting impurities	Limit of detection (B) [r]	100.	0.001	0.10
Photon-emitting impurities	Limit of detection (B) [r]	100.	0.001	0.10
Relative Combined Standard Uncertainty of the Output Quantity, $u_c(y)/y$, (%)				0.34
Coverage Factor, k				<u>$\times 2$</u>
Relative Expanded Uncertainty of the Output Quantity, $U(y)$, (%)				0.68

NOTES

- [a] The Sievert is the SI unit for dose equivalent. See reference [1]. One μSv is equal to 0.1 mrem.
 Distance from Ampoule (cm): 1 30 100
 Approximate Dose Rate ($\mu\text{Sv/h}$): <0.1 - -
- [b] **Massic activity** is the preferred name for the quantity activity divided by the total mass of the sample. See reference [1].
- [c] The reported value, y , of massic activity (activity per unit mass) at the reference time was not measured directly but was derived from measurements and calculations of other quantities. This can be expressed as $y = f(x_1, x_2, x_3, \dots, x_n)$, where f is a mathematical function derived from the assumed model of the measurement process.
- The value, x_i , used for each input quantity i has a **standard uncertainty**, $u(x_i)$, that generates a corresponding uncertainty in y , $u(y) = |\partial y / \partial x_i| \cdot u(x_i)$, called a **component of combined standard uncertainty** of y .
- The **combined standard uncertainty** of y , $u_c(y)$, is the positive square root of the sum of the squares of the components of combined standard uncertainty.
- The combined standard uncertainty is multiplied by a **coverage factor** of $k = 2$ to obtain U , the **expanded uncertainty** of y .
- Since it can be assumed that the possible estimated values of the massic activity are approximately normally distributed with approximate standard deviation $u_c(y)$, the unknown value of the massic activity is believed to lie in the interval $y \pm U$ with a level of confidence of approximately 95 percent.
- For further information on the expression of uncertainties, see references [2] and [3].
- [d] The value of each component of combined standard uncertainty, and hence the value of the expanded uncertainty itself, is a best estimate based upon all available information, but is only approximately known. That is to say, the "uncertainty of the uncertainty" is large and not well known. This is true for uncertainties evaluated by statistical methods (e.g., the relative standard deviation of the standard deviation of the mean for the massic count rate is approximately 50%) and for uncertainties evaluated by other methods (which could easily be over estimated or under estimated by substantial amounts). The unknown value of the expanded uncertainty is believed to lie in the interval $U/2$ to $2U$ (i.e., within a factor of 2 of the estimated value).
- [e] The stated uncertainty is two times the standard uncertainty.
- [f] Estimated limits of detection for alpha-particle-emitting impurities are:
 0.04 $\text{s}^{-1} \cdot \text{g}^{-1}$ for energies less than 4.9 MeV and
 0.001 $\text{s}^{-1} \cdot \text{g}^{-1}$ for energies greater than 5.2 MeV.
 From mass-spectrometric measurements performed by the supplier, the massic activities of other detected radionuclides (in $\text{Bq} \cdot \text{g}^{-1}$ at the reference time) are:
 ^{240}Pu : 0.002; ^{241}Pu : ≈ 0.02 ; ^{242}Pu : ≈ 0.000003 ; ^{241}Am : ≈ 0.001
 From the photon measurements below, we have ^{241}Am : ≤ 0.0006
- [g] Estimated limits of detection for photon-emitting impurities are:
 0.000200 $\text{s}^{-1} \cdot \text{g}^{-1}$ for energies between 42.5 and 90 keV,
 0.000080 $\text{s}^{-1} \cdot \text{g}^{-1}$ for energies between 102 and 125 keV,
 0.000030 $\text{s}^{-1} \cdot \text{g}^{-1}$ for energies between 133 and 1456 keV, and
 0.000008 $\text{s}^{-1} \cdot \text{g}^{-1}$ for energies between 1465 and 3500 keV,
 provided that the photons are separated in energy by 4 keV or more from photons emitted in the decay of plutonium-239.

- [h] The stated uncertainty is the standard uncertainty.
- [i] Relative standard uncertainty of the input quantity x_i .
- [j] The relative change in the output quantity y divided by the relative change in the input quantity x_i . If $|\partial y / \partial x_i| \cdot (x_i / y) = 1.0$, then a 1% change in x_i results in a 1% change in y . If $|\partial y / \partial x_i| \cdot (x_i / y) = 0.05$, then a 1% change in x_i results in a 0.05% change in y .
- [k] Relative component of combined standard uncertainty of output quantity y , rounded to two significant figures or less. The relative component of combined standard uncertainty of y is given by $u(y)/y = |\partial y / \partial x_i| \cdot u(x_i)/y = |\partial y / \partial x_i| \cdot (x_i / y) \cdot u(x_i)/x_i$. The numerical values of $u(x_i)/x_i$, $|\partial y / \partial x_i| \cdot (x_i / y)$, and $u(y)/y$, all dimensionless quantities, are listed in columns 3, 4, and 5, respectively. Thus, the value in column 5 is equal to the value in column 4 multiplied by the value in column 3. The input quantities are independent, or very nearly so. Hence the covariances are zero or negligible.
- [m] The relative standard uncertainty of $\lambda \cdot t$ is determined by the relative standard uncertainty of λ (i.e., of the half life). The relative standard uncertainty of t is negligible.
- [n] $|\partial y / \partial x_i| \cdot (x_i / y) = |\lambda \cdot t|$
- [p] The live time is determined by counting the pulses from a gated oscillator.
- [q] Includes the uncertainty in the alpha-particle detection efficiency of the liquid-scintillation cocktail and the uncertainty associated with the finite detection threshold of the counting electronics.
- [r] The standard uncertainty for each undetected impurity that might reasonably be expected to be present is estimated to be equal to the estimated limit of detection for that impurity, i.e. $u(x_i)/x_i = 100\%$. $|\partial y / \partial x_i| \cdot (x_i / y) = \{(\text{response per Bq of impurity})/(\text{response per Bq of plutonium-239})\} \cdot ((\text{Bq of impurity})/(\text{Bq of plutonium-239}))$. Thus $u(y)/y$ is the relative change in y if the impurity were present with a massic activity equal to the estimated limit of detection.

REFERENCES

- [1] International Organization for Standardization (ISO), *ISO Standards Handbook- Quantities and Units*, 1993. Available from the American National Standards Institute, 11 West 42nd Street, New York, NY 10036, U.S.A. 1-212-642-4900.
- [2] International Organization for Standardization (ISO), *Guide to the Expression of Uncertainty in Measurement*, 1993. Available from the American National Standards Institute, 11 West 42nd Street, New York, NY 10036, U.S.A. 1-212-642-4900. (Listed under ISO miscellaneous publications as "ISO Guide to the Expression 1993".)
- [3] B. N. Taylor and C. E. Kuyatt, *Guidelines for Evaluating and Expressing the Uncertainty of NIST Measurement Results*, NIST Technical Note 1297, 1994. Available from the Superintendent of Documents, U.S. Government Printing Office, Washington, DC 20407, U.S.A.
- [4] National Council on Radiation Protection and Measurements Report No. 58, *A Handbook of Radioactivity Measurements Procedures*, Second Edition, 1985. Available from the National Council on Radiation Protection and Measurements, 7910 Woodmont Avenue, Bethesda, MD 20814 U.S.A.
- [5] Evaluated Nuclear Structure Data File (ENSDF), March 2000.