



National Institute of Standards & Technology

Certificate

Standard Reference Material 4404H, Lot 25 Thallium-201 Radioactivity Standard

This Standard Reference Material (SRM) consists of radioactive thallium-201 nitrate, non-radioactive thallium 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 ionization chambers and solid-state gamma-ray spectrometry systems.

Radiological Hazard

The SRM ampoule contains thallium-201 with a total activity of approximately 500 MBq. Thallium-201 decays by electron capture. During the decay process, X-rays and gamma rays with energies from approximately 2 keV to 167 keV are emitted. Most of these photons escape from the SRM ampoule and can represent a radiation hazard. Approximate unshielded dose rates at several distances (as of the reference time) are given in note [a]*. Appropriate shielding and/or distance should be used to minimize personnel exposure. The SRM should be used only by persons qualified to handle radioactive material.

Chemical Hazard

The SRM ampoule contains nitric acid with a concentration of approximately 1 mole 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 June 2003. 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) both 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, B.M. Coursey, Acting Group Leader. The overall technical direction and physical measurements leading to certification were provided by D.B. Golas and O.T. Palabrica, Nuclear Energy Institute Research Associates. 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.

Bert M. Coursey, Chief
Ionizing Radiation Division

Gaithersburg, Maryland 20899
August 2002

John Rumble, Jr., Chief
Measurement Services Division

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 4404H, Lot 25, Ampoule 1

Certified values

Radionuclide	Thallium-201
Reference time	1400 EST, 25 June 2002
Massic activity of the solution [b]	95.53 MBq·g⁻¹
Relative expanded uncertainty (k=2)	0.78% [c] [d]
Solution mass	(mass ± 0.0003) g [e]
Solution density	(1.037 ± 0.002) g·mL⁻¹ at 20 °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	
Chemical Properties:			
Solution composition	Chemical Formula	Concentration (mol·L ⁻¹)	Mass Fraction (g·g ⁻¹)
	H ₂ O	54	0.93
	HNO ₃	1.1	0.07
	TlNO ₃	7 × 10 ⁻⁴	2 × 10 ⁻⁴
	²⁰¹ TlNO ₃	6 × 10 ⁻⁸	2 × 10 ⁻⁸
Radiological Properties:			
Photon-emitting impurities	Tl-200: (54 ± 13) kBq·g ⁻¹ [e] [f] Tl-202: (235 ± 59) kBq·g ⁻¹ [e] [f]		
Half lives used	Thallium-200: (26.1 ± 0.1) h [g] [5] Thallium-201: (3.0423 ± 0.0014) d [g] [s] Thallium-202: (12.23 ± 0.02) d [g] [5] Radium-226: (1600 ± 7) a [g] [5]		
Calibration method and measuring instrument(s)	Pressurized "4π"γ ionization chamber A calibrated using a thallium-201 solution whose activity was determined by the 4π(e+X)-γ-coincidence efficiency-extrapolation technique.		

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

Input Quantity x_i , the source of uncertainty (and individual uncertainty components where appropriate)	Method Used To Evaluate $u(x_i)$, the standard uncertainty of x_i (A) denotes evaluation by statistical methods (B) denotes evaluation by other methods	Relative Uncertainty Of Input Quantity, $u(x_i)/x_i$, (%) [h]	Relative Sensitivity Factor, $ \partial y/\partial x_i \cdot$ (x_i/y) [i]	Relative Uncertainty Of Output Quantity, $u_i(y)/y$, (%) [j]
Dilution of SRM 4404H to make SRM 4404L	Estimated (B)	0.05	1.0	0.05
PIC A net response per gram of SRM 4404L, measured relative to RRS200 [k]	Standard deviation of the mean for > 100 repeated measurements (A)	0.02	1.0	0.02
PIC A net response per Bq of thallium-201 in solution, measured relative to RRS200	Standard deviation of the mean for > 100 repeated measurements (A)	0.01	1.0	0.01
Activity used to calibrate PIC A net response per Bq of thallium-201 in solution	Standard uncertainty of the activity determined by the $4\pi(e+X)$ - γ -coincidence efficiency-extrapolation technique. (B)	0.35	1.0	0.35
Half life of thallium-201 Half life of radium-226	Standard uncertainty of the half life (A)	0.04 [m] 0.44 [m]	0.02 [n] 0.01 [n]	0.001 0.004
Gravimetric measurements	Estimated (B)	0.05	1.0	0.05
Live time [p]	Estimated (B)	0.05	1.0	0.05
PIC A charge collection	Estimated (B)	0.05	1.0	0.05
Source positioning	Estimated (B)	0.05	1.0	0.05
Photon-emitting impurities	Estimated (B) [q] Estimated (B) [q] Limit of detection (B) [r]	14. 12. 100.	0.004 0.006 0.001	0.05 0.07 0.10
Relative Combined Standard Uncertainty of the Output Quantity, $u_c(y)/y$, (%)				0.39
Coverage Factor, k				<u>x 2</u>
Relative Expanded Uncertainty of the Output Quantity, U/y , (%)				0.78

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 ($\mu\text{Sv/h}$): | 10000 | 100 | 10 |
- [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 , $u_i(y) = |\partial y / \partial x_i| \cdot u(x_i)$, 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 standard uncertainty component, 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 response 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 photon-emitting impurities, as of 26 June 2002 (0.5 day after the reference time), expressed as massic photon emission rates, are:
- $3 \times 10^4 \text{ s}^{-1} \cdot \text{g}^{-1}$ for energies between 40 keV and 85 keV,
 - $4 \times 10^3 \text{ s}^{-1} \cdot \text{g}^{-1}$ for energies between 86 keV and 163 keV,
 - $1 \times 10^3 \text{ s}^{-1} \cdot \text{g}^{-1}$ for energies between 172 keV and 250 keV,
 - $7 \times 10^2 \text{ s}^{-1} \cdot \text{g}^{-1}$ for energies between 251 keV and 3000 keV, provided that the photons are separated in energy by 4 keV or more from photons emitted in the decay of thallium-201.
- [g] The stated uncertainty is the standard uncertainty.
- [h] Relative standard uncertainty of the input quantity x_i .
- [i] 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 .

- [j] Relative component of combined standard uncertainty of output quantity y , rounded to two decimal places. The relative component of combined standard uncertainty of y is given by $u_i(y)/y \equiv |\partial y/\partial x_i| \cdot u(x_i)/y = |\partial y/\partial x_i| \cdot (x_i/y) \cdot u(x_i)/x_i$. The numerical values of $u(x_i)/x_i$, $|\partial y/\partial x_i| \cdot (x_i/y)$, and $u_i(y)/y$, 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.
- [k] The response of pressurized ionization chamber A (PIC A) is determined from measurement of the time required to collect a given amount of charge on a stable fixed capacitor. All of the response measurements in the NIST pressurized ionization chambers are made relative to the response of one or more artifact standards. These artifact standards consist of microgram quantities of aged radium-226 in small welded stainless-steel capsules. These capsules are encapsulated in plastic rods whose dimensions are similar to those of the standard NIST ampoule. The artifact standards are called **Radium Reference Sources** and are designated as RRS x , where x is the nominal mass (in micrograms) of radium-226 in the capsule.
- [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 crystal-controlled oscillator.
- [q] The standard uncertainties given are for the detected impurities. $|\partial y/\partial x_i| \cdot (x_i/y) = \{(\text{response per Bq of impurity})/(\text{response per Bq of Tl-201})\} \cdot \{(\text{Bq of impurity})/(\text{Bq of Tl-201})\}$.
- [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. $u(x_i)/x_i = 100\%$. $|\partial y/\partial x_i| \cdot (x_i/y) = \{(\text{response per Bq of impurity})/(\text{response per Bq of Tl-201})\} \cdot \{(\text{Bq of impurity})/(\text{Bq of Tl-201})\}$. Thus $u_i(y)/y$ is the relative change in y if the impurity were present with a massic activity equal to the estimated limit of detection.
- [s] This is the equally-weighted mean and the standard deviation of the mean of five published half-life values for thallium-201:

	Published Half Life and One Standard Uncertainty	Standard Deviations From the Mean
1.	(3.0456 ± 0.0015) days [6]	+1.1
2.	(3.043 ± 0.003) days [7]	+0.3
3.	(3.0408 ± 0.0013) days [8]	-0.5
4.	(3.0379 ± 0.0007) days [9]	-1.5
5.	(3.0440 ± 0.0048) days [9]	+0.6

Mean (3.0423 ± 0.0014) days = (73.015 ± 0.015) hours

The value currently recommended in the Evaluated Nuclear Structure Data File (ENSDF) is based upon only value number 4 above. We believe that all of the above data should be considered and that a larger uncertainty is appropriate.

REFERENCES

- [1] International Organization for Standardization (ISO), *ISO Standards Handbook - Quantities and Units*, 1993. Available from Global Engineering Documents, 12 Inverness Way East, Englewood, CO 80112, U.S.A. Telephone 1-800-854-7179.
- [2] International Organization for Standardization (ISO), *Guide to the Expression of Uncertainty in Measurement*, 1993 (corrected and reprinted, 1995). Available from Global Engineering Documents, 12 Inverness Way East, Englewood, CO 80112, U.S.A. Telephone 1-800-854-7179.
- [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), August 2002.
- [6] M.P. Unterweger, D.D. Hoppes and F.J. Schima, New and Revised Half-Life Measurements Results, *Nuclear Instruments and Methods in Physical Research* **A312** (1992) 349.
- [7] H. Schrader, Measurement of the Half-lives of ^{18}F , ^{56}Co , ^{125}I , ^{195}Au and ^{201}Tl , *Applied Radiation and Isotopes* **40** (1989) 383.
- [8] F. Lagoutine and J. Legrand, Périodes de Neuf Radionucléides, *International Journal of Applied Radiation and Isotopes* **33** (1982) 711.
- [9] K. Debertain et al., Decay Data of ^{67}Ga and ^{201}Tl , *International Journal of Applied Radiation and Isotopes* **30** (1979) 551.