

# National Institute of Standards & Technology Certificate

# Standard Reference Material 4968 Radon-222 Emanation Standard

This Standard Reference Material (SRM) consists of a polyethylene-encapsulated radium-226 solution that emanates a known quantity of radon-222 gas under specified conditions. The SRM capsule is packaged in, and is intended to be stored in, a 20-mL glass vial with a screw-on cap. The solution contained in the capsule consists of radioactive radium-226 chloride, non-radioactive barium chloride, and hydrochloric acid dissolved in distilled water. The SRM is intended for the calibration of radon-222 measurement systems.

#### Radiological Hazard

The SRM capsule contains radium-226 with a total activity of approximately 500 Bq. Radium-226 decays by alpha-particle emission to gaseous radon-222 which decays by alpha-particle emission. The progeny of radon-222 decay by alpha-particle and beta-particle emission. None of the alpha particles escape from the SRM capsule, but some of the beta particles escape. During the decay processes, X-rays and gamma rays with energies from 8 keV to 1.8 MeV are emitted. Most of these photons escape from the SRM capsule but their intensities are so small that they do not represent a radiation hazard. Approximate unshielded dose rates for the capsule (in its storage vial) at several distances are given in note [a]\*. Gaseous radon-222 continuously emanates from the SRM capsule and it and its radioactive progeny accumulate within the storage vial. The storage vial should be opened, and the SRM capsule used, only by persons qualified to handle radioactive material.

#### **Chemical Hazard**

This SRM is designed to be used as a sealed capsule and should not be opened. The sealed capsule contains hydrochloric acid (HCl) with a concentration of 1.4 moles per liter of water. The solution is corrosive and represents a health hazard if it comes in contact with eyes or skin. The capsule should be handled only by persons qualified to work with radioactive material and strong acid solution.

#### Handling

The SRM capsule itself is not marked. Labeling and markings are on the storage vial. The storage vial (or any subsequent container) for the SRM capsule should always be clearly marked as containing radioactive material. If it is transported, it should be packed, marked, labeled, and shipped in accordance with the applicable national, international, and carrier regulations. The solution in the capsule is a dangerous good (hazardous material) both because of the radioactivity and because of the strong acid content.

#### Preparation

This SRM 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 R. Collé 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.

Gaithersburg, Maryland 20899 August 1999 Thomas E. Gills, Chief Standard Reference Materials Program

#### Storage

At all times when not in use, the SRM capsule should be stored in water-saturated air at a temperature between 10 and 40 °C. This may be accomplished by storing the capsule in the original storage vial or in any other suitable closed container in which the capsule is either immersed in water or suspended above water. The latter condition is believed to be more effective in rapidly achieving a pre-conditioning "equilibrium" before each use. See the *Information for Users of SRM 4968*.

If properly stored and used, the radon-222 emanation fraction should remain stable until at least September 2004.

#### Additional Information About the SRM Capsule

Radium-226, with a half-life of approximately 1600 years, decays by alpha-particle emission to gaseous radon-222 which decays by alpha-particle emission. The relatively short-lived progeny of radon-222 (polonium-218, lead-214, bismuth-214, and polonium-214) decay by alpha-particle and beta-particle emission to lead-210, which has a half-life of approximately 22 years. Lead-210 decays by beta-particle emission to radioactive bismuth-210 which decays by beta-particle emission to polonium-210. Polonium-210 decays by alpha-particle emission to stable lead-206. The capsule contains varying quantities of each of the above radionuclides. The radium-226 in the capsule was chemically purified many decades ago and, due to ingrowth since that time, the lead-210 activity is now about 80 percent of the radium-226 activity.

The SRM is certified in terms of two parameters that allow calculation of an accumulated radon-222 activity:

- (1) the radium-226 activity in the capsule and
- (2) the radon-222 emanation fraction.

The radon-222 emanation fraction is defined as the fraction of the total radon-222 generated by decay of radium-226 that is released from the capsule and contained within the volume of an accumulation vessel.

The Information for Users of SRM 4968 provides further information on the proper use of the SRM capsule.

For general information about handling and measuring radioactive material, see also reference [4].

# PROPERTIES OF SRM 4968 (Certified values are shown in bold type)

Source identification number	NIST SRM 4968-Number ~					
Physical Properties:						
Source description	Liquid in heat-sealed polyethylene capsule					
Capsule specifications	Right circular cylinder of low-density polyethylene.  Nominal inside diameter of polyethylene = 0.34 cm  Nominal outside diameter of polyethylene = 0.45 cm  Nominal inside length of polyethylene = 1.7 cm  Nominal outside length of polyethylene = 2.0 cm  Nominal polyethylene mass = 0.19 g  Nominal solution mass = 0.14 g					
Total capsule mass	(Mass~ ± 0.002) g	[b] <sup>-</sup>				
Chemical Properties:						
Solution composition	Chemical Formula	Concentration (mol•L <sup>-1</sup> )	Mass Fraction (g•g <sup>-1</sup> )			
	H <sub>2</sub> O HCl BaCl <sub>2</sub> <sup>226</sup> RaCl <sub>2</sub>	54 1.4 0.008 6 × 10 <sup>-8</sup>	$0.95$ $0.05$ $0.002$ $2 \times 10^{-8}$			
Radiological Properties:	Radiological Properties:					
Radionuclide	Radium-226 / Radon-222					
Reference time	1200 EST, 15 September 1998					
Radium-226 activity	Activity∼ Bq					
Relative expanded uncertainty $(k=2)$ of the radium-226 activity	1.4% [d] [e]					
Radon-222 emanation fraction [c]	0.884 at 21°C					
Relative expanded uncertainty $(k=2)$ of the radon-222 emanation fraction	5.2% [d] [e]					
Half lives used	Radon-222: (3.8235 ± 0.0003) d [f] [5] Radium-226: (1600 ± 7) a [f] [5]					
Measuring instrument(s) and calibration method for the radium-226 activity	Pressurized "4π"γ ionization chamber "A" calibrated using national radium standards, liquid scintillation spectrometry, and pulse ionization chamber radon analyses. [g]					
Measuring instrument(s) and calibration method for the radon-222 emanation fraction	Pulse ionization chambers (part of the NIST primary radon measurement system [6,7]) calibrated using national radium standards [g].					

## EVALUATION OF THE UNCERTAINTY OF THE RADIUM-226 ACTIVITY [d]\*

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 $ $(x_i/y)$ [i]	Relative Uncertainty Of Output Quantity, $u_i(y)/y$ , $(\%)$ [j]
Pressurized ionization chamber "A" net response per gram of SRM 4968 solution, measured relative to eight "1947 (1967 recalibrated) series" of radium-226 standards [g]	Standard deviation of the mean for 24 sets of ionization-chamber measurements (A)	0.08	1.0	0.08
Calibration of the "1947 (1967 recalibrated) series" of radium-226 standards [g]	Estimated (B)	0.34	1.0	0.34
Conversion of radium- 226 mass to activity [k]	Standard uncertainty of the half life of radium-226 (A)	0.44	1.0	0.44
Corrections for decay of radium-226 [m]	Standard uncertainty of the half life of radium-226 (A)	0.44	0.004 [n]	0.002
Gravimetric measurements	Estimated (B)	0.25	1.0	0.25
Live time [p]	Estimated (B)	0.05	1.0	0.05
Charge collection efficiency	Estimated (B)	0.05	1.0	0.05
Source Positioning	Estimated (B)	0.3	1.0	0.3
Photon-emitting impurities	Limit of detection (B) [q]	100.	0.0001	0.01
Relative Combined Standard Uncertainty of the Output Quantity, $u_c(y)/y$ , (%)				
Coverage Factor, k				
Relative Expanded Uncertainty of the Output Quantity, U/y, (%)				

## EVALUATION OF THE UNCERTAINTY OF THE RADON-222 EMANATION FRACTION [d]\*

			r	
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 $ $(x_i/y)$ [i]	Relative Uncertainty Of Output Quantity, u <sub>i</sub> (y)/y, (%) [j]
Pulse ionization chamber extrapolated net count rate per Bq of radium-226 in the capsule, corrected for radon-222 accumulation time, decay, transfer efficiency, and detection efficiency	Typical standard deviation of the mean for measurements repeated weekly (for 3 to 6 weeks) on a single capsule (A)  Standard deviation of the mean for 11 repeated measurements over 6 years on a single capsule (A)	0.7	1.0	0.7
	Standard deviation of the distribution of the mean values obtained for 32 capsules (A)	2.4	1.0	2.4
Live time	Estimated (B)	0.1	1.0	0.1
Extrapolation of net- count-rate-versus-energy to zero energy	Estimated (B)	0.3	1.0	0.3
Corrections for accumulation and decay of radon-222 [r]	Estimated (B)	0.09	1.0	0.09
Corrections for decay of radium-226 [m]	Estimated (B)	0.44	0.002 [n]	0.001
Transfer efficiency for radon-222	Estimated (B)	0.2	1.0	0.2
Ratio of the radium-226 activity in the capsule to that in a standardized solution used to calibrate the pulse ionization chamber	Estimated (B)	0.38	1.0	0.38
			1.0	
Relative Combined Standard Uncertainty of the Output Quantity, $u_c(y)/y$ , (%)				2.6 x 2
Coverage Factor, k				
Relative Expanded Uncertainty of the Output Quantity, U/y, (%)				

#### **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 (μSv/h): <1 -
- [b] The stated uncertainty is two times the standard uncertainty. The SRM should not be used if the total mass of the capsule decreases by more than 0.070 grams. See the *Information for Users of SRM* 4968.
- [c] The emanation fraction is defined as the fraction of the total radon-222 generated by decay of radium-226 during a given accumulation time interval that is released from the capsule and contained within the volume of an accumulation vessel. See the *Information for Users of SRM 4968*.
- [d] The reported value, y, of the radium-226 activity or of the radon-222 emanation fraction 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].

- [e] 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).
- [f] The stated uncertainty is the standard uncertainty.
- [g] For further details on the calibration of the various NIST (NBS) radium series see reference [8]. The 1967 recalibrations of the "1947 series" and of the "1957 series" were made using pressurized " $4\pi$ " $\gamma$  ionization chamber "A". The radium-226 solutions used for the present calibrations, and for filling the capsules, were directly compared to the "1992 series" of radium standards (SRM 4965, 4966, and 4967) by liquid scintillation counting. The latter SRMs were directly compared against the "1947 (1967 recalibration)", "1978", and "1984" series by radon-222 analyses with the pulse ionization chambers, by liquid scintillation counting, and by NaI(TI) well-crystal and Ge  $\gamma$  spectrometry.

- [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.
- 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  $u_i(y)/y = |\partial y/\partial x_i| \cdot u(x_i)/y = |\partial y/\partial x_i| \cdot (x_i/y) \cdot u(x_i)/x_i$ . The numerical values of  $u(x_i)/x_i$ ,  $|\partial y/\partial x_i| \cdot (x_i/y)$ , and  $u_i(y)/y$ , 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 relative standard uncertainty of the activity per unit mass of radium-226 is determined by the relative standard uncertainty of  $\lambda$  (i.e., of the half life). The relative standard uncertainty of the atomic weight of radium-226, and of Avogadro's number, is negligible.
- [m] The relative standard uncertainty of  $\lambda \cdot t$  is determined by the relative standard uncertainty of  $\lambda$  (i.e., of the half life). The relative standard uncertainty of t is negligible.
- [n]  $\left| \frac{\partial y}{\partial x_i} \right| \cdot (x_i/y) = \left| \lambda \cdot t \right|$
- [p] The live time is determined by counting the pulses from a gated oscillator.
- [q] 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.  $u(x_i)/x_i = 100\%$ .  $|\partial y/\partial x_i| \cdot (x_i/y) = \{(\text{response per Bq of impurity})/(\text{response per Bq of Ra-226})\} \cdot \{(\text{Bq of impurity})/(\text{Bq of Ra-226})\}$ . 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.
- [r] The relative standard uncertainty of both the accumulation correction and the decay correction for radon-222 is largely determined by the (approximately 0.5 %) relative standard uncertainty of the time for the transfer of the accumulated radon into the pulse ionization chambers. The relative standard uncertainty of the radon-222 half life is negligible.

#### 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.
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- [5] Evaluated Nuclear Structure Data File (ENSDF), July 1999.
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- [8] W.B. Mann, L.L. Stockman, W.J. Youden, A. Schwebel, P.A. Mullen and S.B. Garfinkel, Preparation of New Solution Standards of Radium, *J. Res. Natl. Bur. Stds.* 62, 21-26 (1959).