



## Standard Test Method for Radium-226 in Water<sup>1</sup>

This standard is issued under the fixed designation D 3454; the number immediately following the designation indicates the year of original adoption or, in the case of revision, the year of last revision. A number in parentheses indicates the year of last approval. A superscript epsilon ( $\epsilon$ ) indicates an editorial change since the last revision or reapproval.

### 1. Scope

1.1 This test method covers the measurement of soluble, suspended, and total radium-226 in water in concentrations above  $3.7 \times 10^{-3}$  Bq/L. This test method is not applicable to the measurement of other radium isotopes.

1.2 This test method may be used for quantitative measurements by calibrating with a radium-226 standard, or for relative measurements by comparing the measurements made with each other.

1.3 This test method does not meet the current requirements of Practice D 2777.

1.4 The values stated in SI units are to be regarded as the standard. The inch-pound units given in parentheses are for information only.

1.5 Hydrofluoric acid (HF) is very hazardous and should be used in a well-ventilated hood. Wear rubber gloves, safety glasses or goggles, and a laboratory coat. Avoid breathing any HF fumes. Clean up all spills promptly and wash thoroughly after using HF.

1.6 *This standard does not purport to address all of the other safety concerns, if any, associated with its use. It is the responsibility of the user of this standard to establish appropriate safety and health practices and determine the applicability of regulatory limitations prior to use.*

### 2. Referenced Documents

#### 2.1 ASTM Standards:

D 1129 Terminology Relating to Water<sup>2</sup>

D 1193 Specification for Reagent Water<sup>2</sup>

D 2777 Practice for Determination of Precision and Bias Applicable Methods of Committee D-19 on Water<sup>2</sup>

D 3370 Practices for Sampling Water from Closed Conduits<sup>2</sup>

D 3649 Practice for High-Resolution Gamma-Ray Spectrometry of Water<sup>3</sup>

<sup>1</sup> This test method is under the jurisdiction of ASTM Committee D 19 on Water and is the direct responsibility of Subcommittee D19.04 on Methods of Radiochemical Analysis.

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<sup>2</sup> *Annual Book of ASTM Standards*, Vol 11.01.

<sup>3</sup> *Annual Book of ASTM Standards*, Vol 11.02.

### 3. Terminology

3.1 *Definitions*—For definitions of terms used in this test method, refer to Terminology D 1129, and to other published glossaries.<sup>4</sup>

### 4. Summary of Test Method

4.1 This test method<sup>5</sup> is based on the emanation and scintillation counting of radon-222, a gaseous daughter product of radium-226, from a solution.

4.2 Radium-226 is collected from water by coprecipitation on a relatively large amount of barium sulfate. The barium-radium sulfate is decomposed by fuming with phosphoric acid, and the resulting glassy melt is dissolved by evaporation with dilute hydrochloric acid to form soluble barium-radium phosphates and chlorides. These salts are dissolved and the solution is stored for ingrowth of radon-222. After a suitable ingrowth period, the radon gas is removed from the solution by purging with gas and transferred to a scintillation counting chamber. About 4 h after radon-222 collection, the scintillation chamber is counted for alpha activity. The radium-226 concentration is calculated from the alpha count rate of radon-222 and its immediate daughters. The radioactive decay characteristics of radium-226 and its immediate decay progeny are listed in Table 1.

### 5. Significance and Use

5.1 The most prevalent of the five radium isotopes in ground water, having a half life greater than one day, are radium-226 and radium-228. These two isotopes also present the greatest health risk compared to the other naturally occurring nuclides of equal concentrations if ingested via the water pathway.

5.2 Although primarily utilized on a water medium, this technique may be applicable for the measurement of the radium-226 content of any media once the medium has been completely decomposed and put into an aqueous solution.

<sup>4</sup> American National Standard Glossary of Terms in Nuclear Science and Technology, N1.1-1967.

<sup>5</sup> This test method is based on a previously published method by Rushing, D.E., Garcia, W.J., and Clark, D.A. "The Analysis of Effluents and Environmental Samples from Uranium Mills and of Biological Samples for Radium, Polonium and Uranium," Radiological Health and Safety in Mining and Milling of Nuclear Materials, Vol. II, IAEA, Vienna, Austria, 1964), p. 187.

**TABLE 1 Radioactive Decay Characteristics of Radium-226 and Its Daughters**

Radionuclide	Half-life	Mode of Decay
Radium-226	1602 years	$\alpha$
Radon-222	3.82 days	$\alpha$
Polonium-218	3.05 min	$\alpha$
Lead-214	26.8 min	$\beta, \gamma$
Bismuth-214	19.7 min	$\beta, \gamma$
Polonium-214	164 $\mu$ s	$\alpha$
Lead-210	22.3 years	$\beta, \gamma$

5.3 The general methodology and basis of this technique are similar to the methodology "Radium-226 in Drinking Water (Radon Emanation Technique)" as described in the document EPA-600/4-80-032.<sup>6</sup>

**6. Interferences**

6.1 Only the gaseous alpha-emitting radionuclides interfere, namely, radon-219 and radon-220. Their half lives are 3.9 and 54.5 s respectively; their presence indicates the presence of their parents, radium-223 and radium-224. These short-lived radon isotopes decay before the radon-222 is counted; it is their alpha-emitting decay products that would interfere. These interferences are very rare in water samples but are frequently observed in certain uranium mill effluents.

**7. Apparatus**

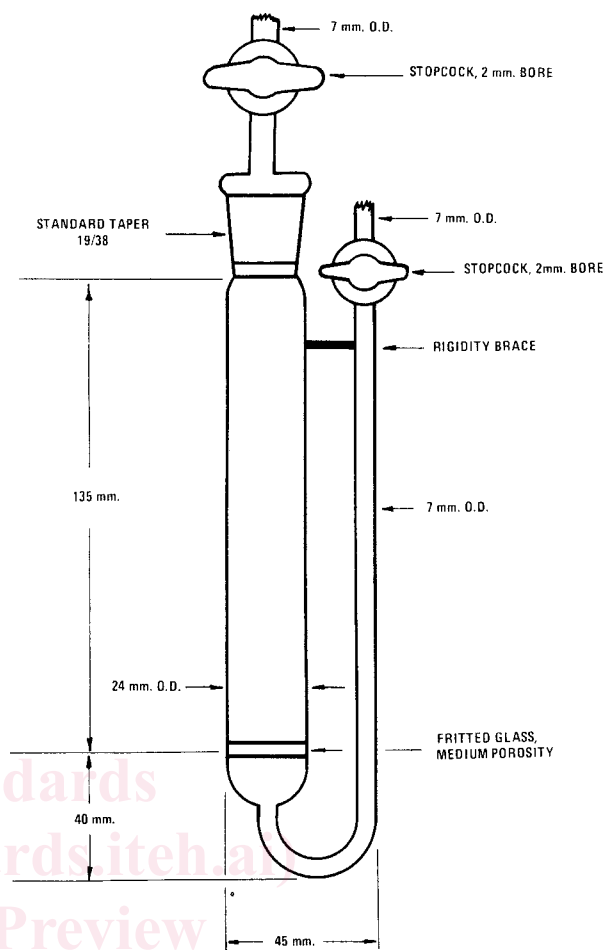
7.1 *Radon Bubbler*<sup>7</sup> (Fig. 1).

7.2 *Radon Scintillation Chamber*<sup>8</sup> (also known as Lucas Cell) (Fig. 2).

7.3 *Manometer*, open-end capillary tube or vacuum gage having a volume which is small compared to the volume of the scintillation chamber, 0, - 760 mm Hg (Fig. 3).

7.4 *Gas Purification Tube*, 7 to 8 mm outside diameter standard wall glass tubing, 100 mm long, constricted at lower end to hold a glass wool plug (Fig. 3). The upper half of the tube is filled with magnesium perchlorate and the lower half with a sodium hydrate-asbestos absorbent.

7.5 *Scintillation Counter Assembly*, consisting of a 50 mm (2 in.) or more in diameter photomultiplier tube mounted in a light-tight housing and coupled to the appropriate preamplifier, high-voltage supply, and scaler. A high-voltage safety switch should open automatically when the light cover is removed to avoid damage to the photomultiplier tube. The preamplifier should incorporate a variable gain adjustment. The counter should be equipped with a flexible ground wire which is attached to the chassis photomultiplier tube by means of an alligator clip or similar device. The operating voltage is



**FIG. 1 Radon Bubbler**

ascertained by determining a plateau using radon-222 in the scintillation chamber as the alpha source. The slope of the plateau should not exceed 2%/100 V. The counter and the scintillation chamber should be calibrated and used as a unit when more than one counter is available. The background counting rate for the counter assembly without the scintillation chamber should range from 0.00 to 0.03 cpm.

7.6 *Membrane Filters*, 0.45- $\mu$ m pore size.<sup>9</sup>

7.7 *Silicone Grease*, high-vacuum, for bubbler stopcocks.

7.8 *Platinum Ware*, crucibles, 20 to 30 mL, and one 500-mL capacity dish. Platinum ware is cleaned by immersing and rotating in a molten bath of potassium pyrosulfate, removing, cooling, and rinsing in hot tap water, digesting in hot HCl (1+1), rinsing in water, and finally flaming over a burner.

7.9 *Laboratory Glassware*—Glassware may be decontaminated before and between uses by heating for 1 h in EDTA-Na<sub>2</sub>CO<sub>3</sub> decontaminating solution at 90 to 100°C, then rinsing in water, in (1+1) HCl and again in water.

<sup>6</sup> "Radium-226 in Drinking Water (Radon Emanation Technique)," *Prescribed Procedures for Measurement of Radioactivity in Drinking Water*, August 1980.

<sup>7</sup> Available from Corning Glass Works, Special Sales Section, Corning, N.Y. 11830.

<sup>8</sup> Available from W. H. Johnston Laboratories, 3617 Woodland Ave., Baltimore, MD 21215, and Rocky Mountain Scientific Glass Blowing Co., 4990 E. Asbury Ave. Denver, CO 80222.

<sup>9</sup> Type HAWP (Millipore filter Corp., Bedford, MA) has been found satisfactory. An equivalent may be used.

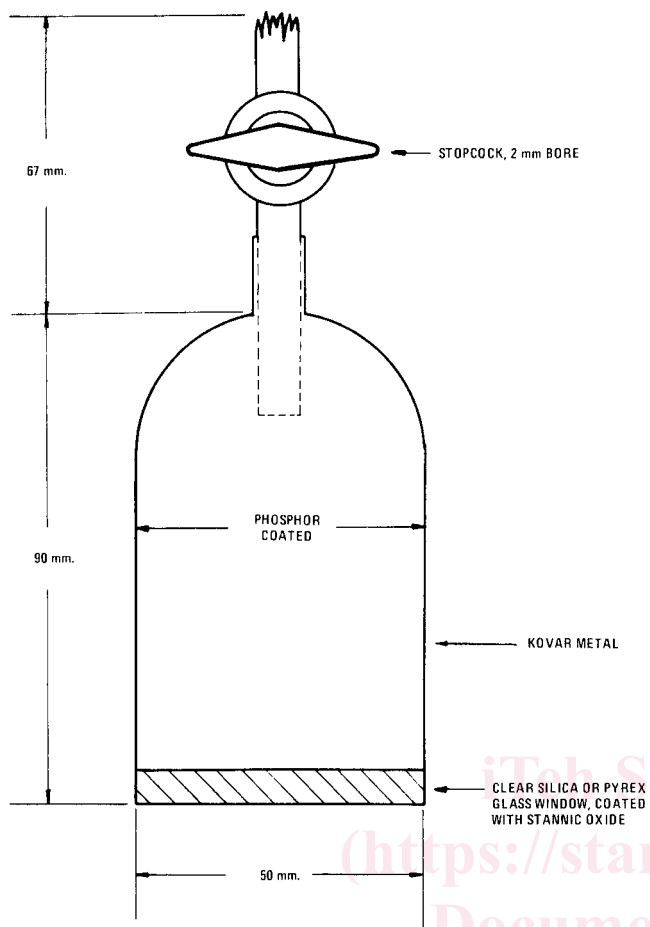


FIG. 2 Radon Scintillation Chamber

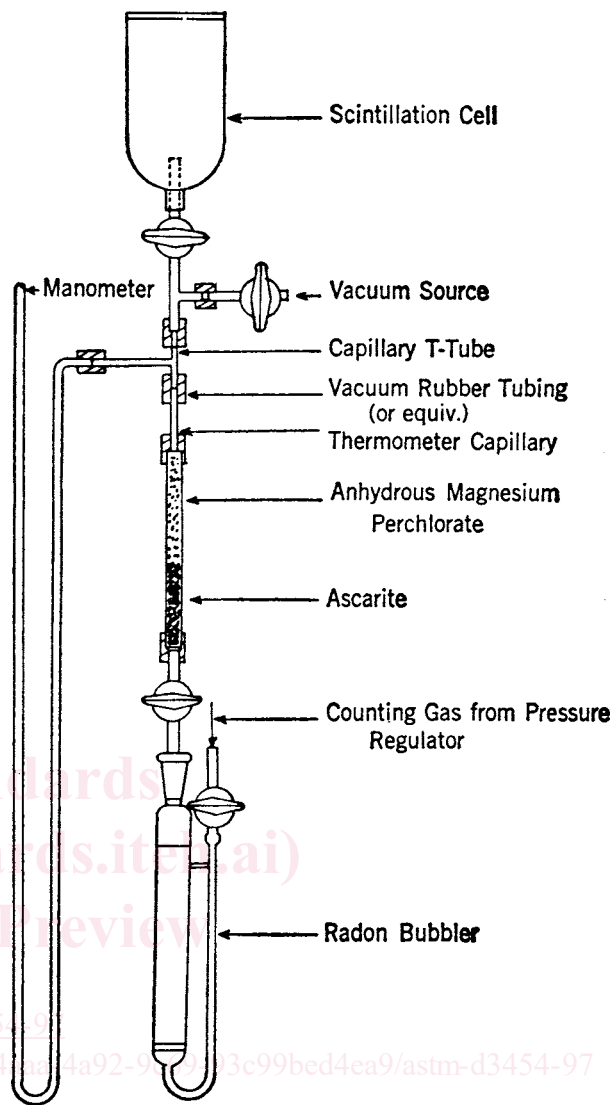


FIG. 3 De-emanation Assembly

## 8. Reagents and Materials

8.1 *Purity of Reagents*—Reagent grade chemicals shall be used in all tests. Unless otherwise indicated, it is intended that all reagents shall conform to the specifications of the Committee on Analytical Reagents of the American Chemical Society.<sup>10</sup> Other grades may be used, provided it is first ascertained that the reagent is of sufficiently high purity to permit its use without lessening the accuracy of the determination.

8.2 *Purity of Water*—Unless otherwise indicated, reference to water shall be understood to mean conforming to Specification D 1193, Type III.

8.3 *Radioactive Purity of Reagents*—Radioactive purity shall be such that the measured results of blank samples do not exceed the calculated probable error of the measurement or are within the desired precision.

8.4 *Ammonium Sulfate Solution (100 g/L)*—Dissolve 10 g of ammonium sulfate ( $(\text{NH}_4)_2\text{SO}_4$ ) in water and dilute to 100 mL.

8.5 *Barium Chloride Carrier Solution Stock, (17.8 g/L)*—Dissolve 17.8 g of barium chloride ( $\text{BaCl}_2 \cdot 2\text{H}_2\text{O}$ ) in water and dilute to 1 L. This solution will contain 10 mg  $\text{Ba}^{+2}$ /mL.

8.6 *Barium-133 Tracer Solution*—(approximately  $2.74 \times 10^3$  Bq/mL).

8.7 *Barium Chloride Carrier Solution, Working*—Add 100 mL of barium chloride carrier stock solution and 10 mL of barium-133 tracer solution to 890 mL of water and mix thoroughly. This solution will contain approximately 1 g of  $\text{Ba}^{+2}$ /L. Allow to stand for 24 h and filter through a membrane filter.

8.8 *EDTA-Sodium Carbonate Decontaminating Solution*—Dissolve 10 g of disodium ethylenediaminetetraacetate and 10 g of sodium carbonate ( $\text{Na}_2\text{CO}_3$ ) in water and dilute to 1 L.

8.9 *Flux*—To a large platinum dish (about 500-mL capacity) add 30 mg of  $\text{BaSO}_4$ , 65.8 g of  $\text{K}_2\text{CO}_3$ , 50.5 g of  $\text{Na}_2\text{CO}_3$ , and 33.7 g of  $\text{Na}_2\text{B}_4\text{O}_7 \cdot 10 \text{H}_2\text{O}$ . Mix well and heat cautiously until the water is expelled; fuse and mix thoroughly by swirling. Cool flux, grind it in a porcelain mortar to pass a U. S. Standard No. 10 (2.00-mm) (or finer) sieve. Store in an airtight bottle. (Flux can be prepared in smaller batches.)

<sup>10</sup> "Reagent Chemicals, American Chemical Society Specifications," Am. Chemical Soc., Washington, DC. For suggestions on testing of reagents not listed by the American Chemical Society, see "Analytical Standards for Laboratory Chemicals," BDH Ltd., Poole, Dorset, U.K., and the "United States Pharmacopeia," and National Formulary, U.S. Pharmaceutical Convention, Inc. (USPC), Rockville, MD.