



## Standard Test Method for Alpha-Particle-Emitting Isotopes of Radium in Water<sup>1</sup>

This standard is issued under the fixed designation D 2460; 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 separation of dissolved radium from water for the purpose of measuring its radioactivity. Although all radium isotopes are separated, the test method is limited to alpha-particle-emitting isotopes by choice of radiation detector. The most important of these radioisotopes are radium-223, radium-224, and radium-226. The lower limit of concentration to which this test method is applicable is  $3.7 \times 10^{-2}$  Bq/L (1 pCi/L).

1.2 This test method may be used for absolute measurements by calibrating with a suitable alpha-emitting radioisotope such as radium-226, or for relative methods by comparing measurements with each other. Mixtures of radium isotopes may be reported as equivalent radium-226. Information is also provided from which the relative contributions of radium isotopes may be calculated.

1.3 *This standard does not purport to address all of the 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.* For a specific precautionary statement, see Section 9.

### 2. Referenced Documents

#### 2.1 ASTM Standards:

- C 859 Terminology Relating to Nuclear Materials<sup>2</sup>
- D 1129 Terminology Relating to Water<sup>3</sup>
- D 1193 Specification for Reagent Water<sup>3</sup>
- D 1943 Test Method for Alpha Particle Radioactivity of Water<sup>4</sup>
- D 2777 Practice for Determination of Precision and Bias of Applicable Methods of Committee D-19 on Water<sup>3</sup>
- D 3370 Practices for Sampling Water<sup>3</sup>
- D 3454 Test Method for Radium-226 in Water<sup>4</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 12.01.

<sup>3</sup> *Annual Book of ASTM Standards*, Vol 11.01.

<sup>4</sup> *Annual Book of ASTM Standards*, Vol 11.02.

D 3648 Practices for the Measurement of Radioactivity<sup>4</sup>

### 3. Terminology

#### 3.1 Definition:

3.1.1 For definitions of terms used in this standard, see Terminology C 859 and D 1129. For terms not included in these, reference may be made to other published glossaries (1, 2).<sup>5</sup>

### 4. Summary of Test Method

4.1 Radium is collected from the water by coprecipitation with mixed barium and lead sulfates. The barium and lead carriers are added to a solution containing alkaline citrate ion which prevents precipitation until interchange has taken place. Sulfuric acid is then used to precipitate the sulfates, which are purified by nitric acid washes. The precipitate is dissolved in ammoniacal EDTA. The barium and radium sulfates are reprecipitated by the addition of acetic acid, thereby separating them from lead and other radionuclides. The precipitate is dried on a planchet weighed to determine the chemical yield, and alpha-counted to determine the total disintegration rate of alpha-particle-emitting radium isotopes. This procedure is based upon published ones (3, 4).

### 5. Significance and Use

5.1 Radium is one of the most radiotoxic elements. Its isotope of mass 226 is the most hazardous because of its long half-life. The isotopes 223 and 224, although not as hazardous, are of some concern in appraising the quality of water.

5.2 The alpha-particle-emitting isotopes of radium other than that of mass 226 may be determined by difference if radium-226 is measured separately, such as by Test Method D 3454. Note that one finds radium-226 and -223 together in variable proportions (5, 6), but radium-224 does not normally occur with them. Thus, radium-223 often may be determined by simply subtracting the radium-226 content from the total: and if radium-226 and -223 are low, radium-224 may be determined directly. The determination of a single isotope in a mixture is less precise than if it occurred alone.

<sup>5</sup> The boldface numbers in parentheses refer to a list of references at the end of this standard.

## 6. Interferences

6.1 A barium content in the sample exceeding 0.2 mg will cause a falsely high chemical yield.

## 7. Apparatus

7.1 For suitable gas-flow proportional or alpha-scintillation counting equipment, refer to Test Method D 1943.

## 8. Reagents

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, where such specifications are available.<sup>6</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 precision, or increasing the bias, of the determination.

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

8.3 *Radioactivity Purity Of Reagents*—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 *Acetic Acid, Glacial* (sp gr 1.05).

8.5 *Ammonium Hydroxide* (sp gr 0.90)—Concentrated ammonium hydroxide (NH<sub>4</sub>OH).

8.6 *Ammonium Hydroxide (1+1)* Mix 1 volume of concentrated ammonium hydroxide (NH<sub>4</sub>OH, sp gr 0.90) with 1 volume of water.

8.7 *Barium Nitrate Carrier Solution* (10 mg Ba/mL)—Dissolve 1.90 g of barium nitrate (Ba(NO<sub>3</sub>)<sub>2</sub>) in water and dilute to 100 mL.

8.8 *Citric Acid Solution* (350 g/L)—Dissolve 350 g of citric acid (anhydrous) in water and dilute to 1 L.

8.9 *Disodium Ethylenediamine Tetraacetate Solution* (93 g/L)—Dissolve 93 g of disodium ethylenediamine tetraacetate dihydrate in water and dilute to 1 L.

8.10 *Lead Nitrate Carrier Solution* (104 mg Pb/mL)—Dissolve 33.2 g of lead nitrate (Pb(NO<sub>3</sub>)<sub>2</sub>) in water and dilute to 200 mL.

8.11 *Methyl Orange Indicator Solution*—Dissolve 1.0 g of methyl orange in water and dilute to 1 L.

8.12 *Nitric Acid* (sp gr 1.42)—Concentrated nitric acid (HNO<sub>3</sub>).

8.13 *Sulfuric Acid* (1 + 1)—Cautiously add with stirring 1 volume of concentrated sulfuric acid (H<sub>2</sub>SO<sub>4</sub>, sp gr 1.84) to 1 volume of water.

## 9. Safety Precautions

9.1 When diluting concentrated acids, always use safety glasses and protective clothing, and add the acid to the water.

<sup>6</sup> *Reagent Chemicals, American Chemical Society Specifications*, American Chemical Society, Washington, DC. For suggestions on the testing of Reagents not listed by the American Chemical Society, see *Analar Standards for Laboratory Chemicals*, BDN Ltd., Poole, Dorset, U.K., and the *United States Pharmacopeia and National Formulary*, U.S. Pharmaceutical Convention, Inc. (USPC), Rockville, MD.

**TABLE 1 Growth of Alpha Activity into Initially Pure Radium-226**

Time, h	Correction, F
0	1.0000
1	1.0160
2	1.0363
3	1.0580
4	1.0798
5	1.1021
6	1.1238
24	1.4892
48	1.9054
72	2.2525

## 10. Sampling

10.1 Collect the sample in accordance with Practices D 3370 as applicable.

10.2 Sample 1 L, or a smaller volume, provided that it is estimated to contain from 3.7 to 370 Bq (100 to 10 000 pCi) of radium. Add 10 mL of HNO<sub>3</sub>/L of sample.

## 11. Calibration and Standardization

11.1 For absolute counting, the alpha-particle detector must be calibrated to obtain the ratio of count rate to disintegration rate. Use NIST traceable radium-226 standards. Analyze two or more portions of such solution, containing known disintegration rates, in accordance with Section 12. After counting, correct the measured activity for chemical yield, and calculate the efficiency, *E* (see Section 13), as the ratio of the observed counting rate to the known disintegration rate.

## 12. Procedure

12.1 Add to a measured volume of sample 5 mL of citric acid and make alkaline (pH > 7.0) with NH<sub>4</sub>OH. Confirm the alkalinity with pH-indicating paper or strip. Add 2 mL of lead carrier and 1.00 mL of barium carrier, and mix.

12.2 Heat to boiling and add 10 drops of methyl orange pH-indicator solution. With stirring, add H<sub>2</sub>SO<sub>4</sub> (1 + 1) until the solution becomes pink, then add 5 drops more.

12.3 Digest the precipitate with continued heating for 10 min. Let cool and collect the precipitate in a centrifuge tube. When large volumes are handled, collection will be facilitated by first letting the precipitate settle, and then decanting most of the clear liquid. Centrifuge then discard the supernatant liquid.

12.4 Wash the precipitate with 10 mL of HNO<sub>3</sub>, centrifuge, and discard the washings. Repeat this wash the precipitate.

12.5 Dissolve the precipitate in 10 mL of water, 10 mL of EDTA solution, and 4 mL of NH<sub>4</sub>OH (1 + 1). Warm if necessary to effect dissolution.

12.6 Reprecipitate barium sulfate (BaSO<sub>4</sub>) by the dropwise addition of acetic acid, then add 3 drops more. Record the time. Centrifuge, then discard the supernatant liquid. Add 10 mL of water, mix well, centrifuge, and discard the supernatant liquid.

12.7 Clean, flame, cool, and weigh a stainless steel planchet that fits the alpha-particle counter being used. Transfer the precipitate to the planchet with a minimum of water. Dry, flame, and weigh the precipitate to determine the chemical yield.

12.8 Promptly count the planchet in an appropriate alpha-particle counter, recording the time. Reserve the planchet for additional measurements, if desired (see 13.4).