



Designation: D 2460 – 05

## 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 reapproval. 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 <sup>223</sup>Ra, <sup>224</sup>Ra, and <sup>226</sup>Ra. 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 <sup>226</sup>Ra, or for relative methods by comparing measurements with each other. Mixtures of radium isotopes may be reported as equivalent <sup>226</sup>Ra. 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:<sup>2</sup>

- C 859** Terminology Relating to Nuclear Materials
- D 1129** Terminology Relating to Water
- D 1193** Specification for Reagent Water
- D 1943** Test Method for Alpha Particle Radioactivity of Water
- D 2777** Practice for Determination of Precision and Bias of Applicable Methods of Committee D-19 on Water
- D 3370** Practices for Sampling Water from Closed Conduits
- D 3454** Test Method for Radium-226 in Water

- D 3648** Practices for the Measurement of Radioactivity
- D 4448** Guide for Sampling Ground-Water Monitoring Wells
- D 5847** Practice for Writing Quality Control Specifications for Standard Test Methods for Water Analysis
- D 6001** Guide for Direct-Push Water Sampling for Geoenvironmental Investigations

### 3. Terminology

#### 3.1 Definition:

3.1.1 For definitions of terms used in this standard, see Terminologies **C 859** and **D 1129**. For terms not included in these, reference may be made to other published glossaries (**1**, **2**).<sup>3</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.

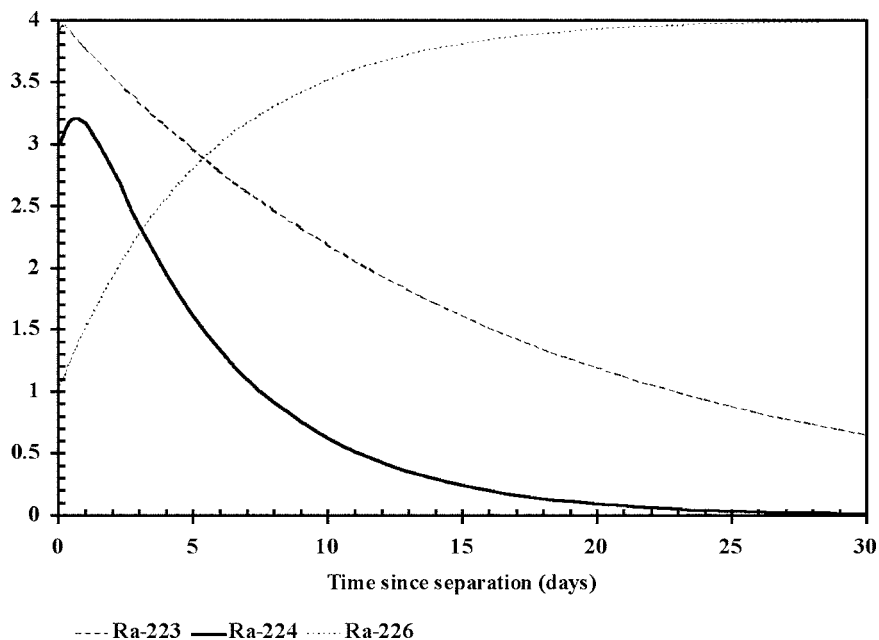
5.2 The alpha-particle-emitting isotopes of radium other than that of mass 226 may be determined by difference if radium-226 is measured separately, such as by Test Method **D 3454**. Note that one finds <sup>226</sup>Ra and <sup>223</sup>Ra together in variable proportions (**5**, **6**), but <sup>224</sup>Ra does not normally occur with them. Thus, <sup>223</sup>Ra often may be determined by simply

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

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<sup>2</sup> For referenced ASTM standards, visit the ASTM website, www.astm.org, or contact ASTM Customer Service at service@astm.org. For *Annual Book of ASTM Standards* volume information, refer to the standard's Document Summary page on the ASTM website.

<sup>3</sup> The boldface numbers in parentheses refer to the list of references at the end of this standard.



NOTE 1—Vertical scale is ratio of the total alpha radioactivity at later time,  $t$ , to radioactivity,  $A_0$ , at initial time of separation.

FIG. 1 Growth and Decay of Alpha Activity into Initially Pure Radium Isotopes

subtracting the  $^{226}\text{Ra}$  content from the total: and if  $^{226}\text{Ra}$  and  $^{223}\text{Ra}$  are low,  $^{224}\text{Ra}$  may be determined directly. The determination of a single isotope in a mixture is less precise than if it occurred alone.

## 6. Interferences

6.1 A barium content in the sample exceeding 0.2 mg will bias chemical yield high and lead to falsely low sample results.

6.2 The presence of suspended solids or insoluble precipitates which fail to dissolve during step 12.5 will bias chemical yield high and lead to falsely low sample results.

6.3 The total alpha particle emission rate from the prepared sample changes over time. This will influence the radium detection efficiency of the counting system used. Initially, the total emission rate will increase as the short-lived radon progeny ingrow in the processed sample. After reaching a maximum, the alpha emission rate will decline at the half life of the radium isotope of interest. In samples of pure isotope, maximum emission rate after radium separation is reached after a period of 4 hours for  $^{223}\text{Ra}$ , 24 hours for  $^{224}\text{Ra}$ , and 28 days for  $^{226}\text{Ra}$ . (See Fig. 1.)

6.4 The alpha particle detection efficiency decreases with increasing precipitate mass. Controlling the precipitate mass relative to that used for calibration of the test will minimize the introduction of significant bias into sample results.

6.5 The changing alpha emission rate and self-absorption effects noted in 6.3 and 6.4 can be addressed by reproducing these conditions during the calibration of the instrument. A series of standards analyzed per 11.2 may be used to generate a curve describing efficiencies over a range of precipitate masses and a series of time encompassing the ingrowth curve (~30 days) of  $^{222}\text{Rn}$  daughters. (See Fig. 2).

## 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>4</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.

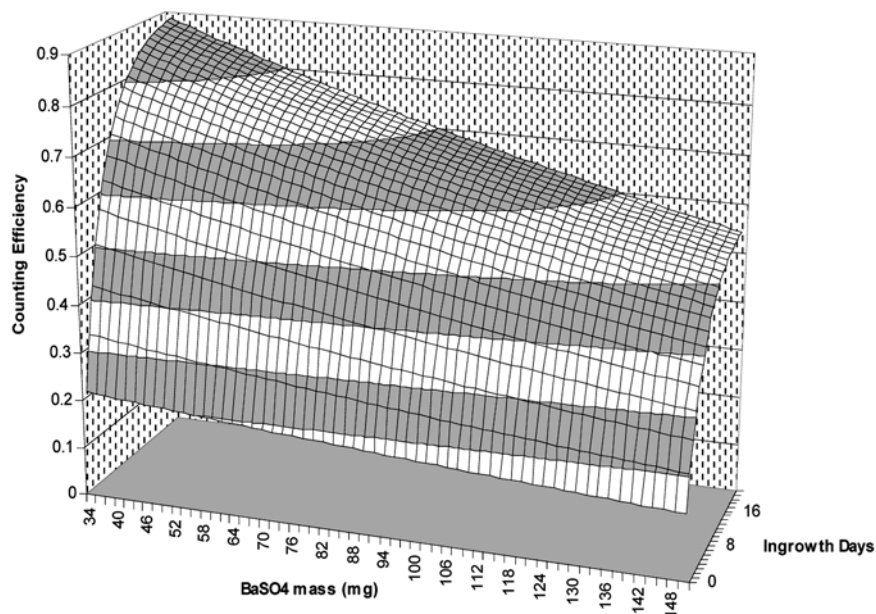
8.3 *Radioactivity Purity of Reagents*, shall be such that the measured results of blank samples do not exceed the calculated probable error of the measurement or are within the desired precision.

8.4 *Acetic Acid, Glacial* (sp gr 1.05).

8.5 *Ammonium Hydroxide* (sp gr 0.90)—Concentrated ammonium hydroxide ( $\text{NH}_4\text{OH}$ ).

8.6 *Ammonium Hydroxide (7 M)*—Mix 1 volume of concentrated ammonium hydroxide ( $\text{NH}_4\text{OH}$ , sp gr 0.90) with 1 volume of water.

<sup>4</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.



Burns, D. C., "Growth and Decay of Alpha Activity into Initially Pure Radium Isotopes," Calibration Plot, Paragon Analytics, Inc., Fort Collins, CO, 2003.

FIG. 2 Typical Alpha Particle Efficiency as Function of Time and Precipitate Mass

8.7 Barium Nitrate Carrier Solution—Standardized (10.0 mg Ba<sup>++</sup>/mL)—Dissolve 1.90 g of barium nitrate (Ba(NO<sub>3</sub>)<sub>2</sub>) in water and dilute to 100 mL.

8.7.1 To perform standardization (in triplicate):

8.7.1.1 Pipette 2.0 mL carrier solution into a centrifuge tube containing 15 mL water.

8.7.1.2 Add 1 mL 18 N H<sub>2</sub>SO<sub>4</sub> while stirring and digest precipitate in a water bath for 10 min.

8.7.1.3 Allow to cool. Centrifuge, and decant supernatant.

8.7.1.4 Wash precipitate with 15 mL water. Centrifuge and decant supernatant.

8.7.1.5 Transfer the precipitate to a tared stainless steel planchet with a minimum of water.

8.7.1.6 Dry under infrared lamp, store in desiccator, and weigh as BaSO<sub>4</sub>.

NOTE 1—1 gram Ba<sup>++</sup> is equivalent to 0.5884 g BaSO<sub>4</sub>

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 (EDTA) (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 (9 M)—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.

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

Time, h	Correction, F
0	1.0000
1	1.0160
2	1.0362
3	1.0578
4	1.0798
5	1.1017
6	1.1235
24	1.4886
48	1.9043
72	2.2513
96	2.5408
120	2.7823
144	2.9839
192	3.2925
240	3.5073
360	3.8006
480	3.9193
720	3.9867

10. Sampling

10.1 Collect the sample in accordance with Practices D 3370, Guide D 4448, or Guide D 6001, 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.

11.2 Use <sup>226</sup>Ra standards traceable to a national standards laboratory (such as NIST or NPL). 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