



Standard Test Method for The Radiochemical Determination of Americium-241 in Soil by Alpha Spectrometry¹

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1. Scope

1.1 This method covers the determination of americium-241 in soil by means of chemical separations and alpha spectrometry. It is designed to analyze up to ten grams of soil or other sample matrices that contain up to 30 mg of combined rare earths. This method allows the determination of americium-241 concentrations from ambient levels to applicable standards. The values stated in SI units are to be regarded as standard.

1.2 *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 specific precaution statements, see Section 9.*

2. Referenced Documents

2.1 ASTM Standards:

C 998 Standard Practice for Sampling Surface Soil for Radionuclides²

C 999 Standard Practice for Soil Sample Preparation for the Determination of Radionuclides²

C 1163 Standard Method for Mounting Actinides for Alpha Spectrometry Using Neodymium Fluoride²

D 1193 Specification for Reagent Water³

D 3084 Standard Practice for Alpha Spectrometry of Water⁴

D 3648 Standard Practices for the Measurement of Radioactivity⁴

3. Summary of Test Method

3.1 Americium-241 is determined in prepared soil samples of up to 10 g. The soil is completely dissolved by use of

pyrosulfate fusion. After an initial separation on barium sulfate and extraction with an organophosphorous compound, the americium is separated from the other trivalent actinides and the rare earths by oxidation of the americium and precipitation of the interferences. The americium is prepared for alpha spectrometry by coprecipitation with neodymium fluoride and the americium-241 determined by alpha spectrometry using americium-243 as a yield monitor.

3.2 Typical radiochemical recoveries of this method as determined by the yield monitor, are between 75 and 90 percent. Decontamination factors from other radionuclides that may interfere with the determination of americium in this energy range are 10^4 – 10^5 .

3.3 The reagent blank contains all reagents plus the americium-243 tracer. Five samples and a reagent blank can be completed and ready for alpha spectrometry in approximately 6 h. The full-width at half-maximum (FWHM) detector resolution ranges between 43 and 65 keV.

4. Significance and Use

4.1 This test method provides the speed and high decontamination factors attainable with liquid-liquid extraction of the actinides and eliminates filtration techniques that are more time consuming.

4.2 This test method provides a precise determination of americium in concentrations normally found in environmental samples.

5. Interferences

5.1 Plutonium, if inadequately separated, may interfere with the alpha spectrometric determination of americium-241. Thorium-228, identifiable by its daughter products, is a serious interference to the final determination of americium by alpha spectrometry if decontamination factors are not sufficiently high. An inadequate separation of polonium-210 may result in an inaccurate determination of the americium-243 yield monitor but this is unlikely when using the neodymium fluoride precipitation method. If high concentrations of these radionuclides are known to be present, a preliminary separation may be required.

¹ This practice is under the jurisdiction of ASTM Committee C26 on Nuclear Fuel Cycle and is the direct responsibility of Subcommittee D26.05 on Methods of Test.

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² Annual Book of ASTM Standards, Vol 12.01.

³ Annual Book of ASTM Standards, Vol 11.01.

⁴ Annual Book of ASTM Standards, Vol 11.02.

6. Apparatus

6.1 Alpha pulse height analysis system as in Practice D 3084.

6.1.1 A system consisting of a silicon surface barrier detector capable of 50 keV or better resolution on standards electrodeposited on a flat, mirror finished disk is required. Samples prepared for alpha spectrometry using neodymium fluoride mounting by ASTM Standard Test Method C 1163 should be capable of 60 to 70 keV resolution. The resolution is defined as the width of an alpha energy peak when the counts on either side of the peak are equal to one-half of the counts at the maximum of the peak (FWHM).

6.1.2 The counting efficiency of the system (i.e., count/disintegration) should be greater than 20 % and the instrument background in the region of each energy peak used for analysis should be less than five counts in 60 000 s (1000 min).

6.2 Membrane Filter (such as cellulose nitrate or cellulose acetate), 47 mm diameter, 0.45 μ m pore size.

7. Reagents and Materials

7.1 *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. 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.

7.2 *Purity of Water*—Unless otherwise indicated, reference to water shall be understood to mean reagent water as defined in Specification D 1193, Type III or better.

7.3 *Americium Tracer*—Purify the americium-243 tracer.⁵ The americium-243 tracer may be available from NIST or other recognized standards laboratories.

7.4 *Anhydrous Potassium Fluoride*.

7.5 *Anhydrous Potassium Sulfate*.

7.6 *Anhydrous Sodium Sulfate*.

7.7 *Ammonium Persulfate* (ammonium peroxydisulfate).

7.8 *Hydrochloric Acid* (sp gr 1.19)—Concentrated hydrochloric acid (HCl).

7.9 *Hydrofluoric Acid* (sp gr 1.20)—Concentrated hydrofluoric acid (HF).

7.10 *Sulfuric Acid* (sp gr 1.84)—Concentrated sulfuric acid (H₂SO₄).

7.11 *0.5 % Sulfuric Acid*—Mix 5 mL of concentrated sulfuric acid with water and dilute to one liter.

7.12 *Nitric Acid* (sp gr 1.42)—Concentrated nitric acid (HNO₃).

7.13 *25 % Potassium Metabisulfite*—Dissolve 25 g of potassium metabisulfite in water and dilute to 100 mL.

7.14 *10 % Sodium Bromate*—Dissolve 10 g of sodium bromate in water and dilute to 100 mL.

7.15 *15 % HDEHP*—Dissolve 150 mL of bis(2-ethylhexyl)phosphoric acid in 850 mL of n-heptane.

7.16 *0.5 % Barium Chloride*—Dissolve 0.5 g of barium chloride in water and dilute to 100 mL.

7.17 *5M Nitric Acid*—Mix 312 mL of concentrated nitric acid with water and dilute to one liter.

7.18 *0.5 % Silver Nitrate*—Dissolve 0.5 g of silver nitrate in water and dilute to 100 mL.

7.19 *Lanthanum Carrier (5 mg La/mL)*—Dissolve 1.17 g of lanthanum nitrate in 75 mL of 5M nitric acid and dilute to 100 mL with water.

7.20 *Phosphoric Acid (sp gr 1.83)*—Concentrated phosphoric acid (H₃PO₄).

7.21 *0.2M Ammonium Persulfate*—Dissolve 2.3 g of ammonium persulfate in water and dilute to 50 mL. Prepare daily.

7.22 *6M Ammonium Fluoride*—Dissolve 22.2 g of ammonium fluoride in water and dilute to 100 mL.

7.23 *0.10M Ammonium Persulfate–3N Ammonium Fluoride*—Mix 20 mL of 0.2M ammonium persulfate with 20 mL of 6M ammonium fluoride. Prepare daily.

7.24 *30 % Hydrogen Peroxide*.

7.25 *Perchloric Acid (sp gr 1.67)*—Concentrated perchloric acid (HClO₄).

7.26 *Neodymium Carrier (10 mg Nd/mL)*—Heat 25 mL of 12N hydrochloric acid and 1.17 g of neodymium oxide on a hot plate until the neodymium oxide is in solution. Cool the solution and dilute to 100 mL with water.

7.27 *Neodymium Carrier (0.5 mg Nd/mL)*—Dilute 5 mL of the 10 mg Nd/mL neodymium carrier solution to 100 mL with water.

8. Sampling

8.1 Collect the sample in accordance with Practice C 998.

8.2 Prepare the sample for analysis in accordance with Practice C 999.

9. Hazards

9.1 In addition to other precautions, adequate laboratory facilities, such as perchloric acid fume hoods and controlled ventilation, along with safe techniques must be used in this procedure. Extreme care should be exercised in using hydrofluoric acid and other hot concentrated acids, particularly hot perchloric acid. Use of safety equipment, especially safety glasses and rubber gloves, is recommended.

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

10. Calibration and Standardization

10.1 If a NIST-Traceable standard americium-243 solution is not available for use as a tracer, standardize a freshly prepared sample of americium-243 using the guidance in Practice D 3648. These practices may also be used to establish the counting efficiency of the alpha spectrometer which then can be used to calculate the chemical recovery.

11. Procedure

11.1 Weigh 10 g of –35 mesh soil, to 0.001 g, into a 250-mL platinum dish and add 30 g of anhydrous potassium fluoride, 10 mL of water, 10 mL of concentrated hydrofluoric acid, and 2 mL of concentrated nitric acid. Slurry the contents of the dish together and add an americium-243 tracer at the level of approximately 0.1 Bq. Place the dish on a fiberglass mat-covered hot plate and evaporate the contents to dryness. Place

⁵ For a description of the process, see Sill, C. W., *Anal. Chem.* 46, 1974, pp. 1426–1431.

the dish on a ring stand and heat with a high temperature blast burner until the contents have dissolved completely. Swirl the contents gently to dissolve any sample on the sides of the dish. When the sample has dissolved completely, carefully remove the platinum dish from the ring stand and swirl the melt as it cools to deposit the melt evenly along the sides and the bottom of the dish.

11.2 After the contents of the dish have cooled to room temperature, carefully add 40 mL of concentrated sulfuric acid to transpose the fluoride cake. After the initial vigorous reaction has subsided, place the dish on a fiberglass mat-covered hot plate and heat until the fluoride cake has been completely transposed. Add 20 g of anhydrous sodium sulfate, place the dish on the ring stand and heat gently until the viscous solution begins to boil. Increase the temperature from the blast burner until a smooth boiling mixture is obtained and continue heating until a clear melt results. Cool the dish to solidify the melt and then place the dish into a cold water bath to facilitate cake removal.

11.3 Bring 500 mL of water and 150 mL of concentrated hydrochloric acid to a boil in a 800-mL beaker and carefully add the contents of the platinum dish to the beaker. Continue boiling until a clear solution results. Add 50 g of anhydrous potassium sulfate and 2 mL of a 25 % potassium metabisulfite solution to the beaker and continue boiling for two minutes.

11.4 To the boiling contents of the beaker, add four 10 mL portions of a 0.5 % barium chloride solution with a 5-minute boiling interval between each addition. Stir the contents of the beaker after each addition of barium chloride. Filter the hot solution through a 47-mm membrane filter using either a glass or polycarbonate filtering chimney. Rinse the beaker and the precipitate with a 0.5 % sulfuric acid solution. Place the filter containing the barium sulfate precipitate into a 125 mL Erlenmeyer flask containing 30 mL of concentrated perchloric acid and heat the contents to fumes of perchloric acid. The use of perchloric acid during the above procedure is used to dissolve the barium sulfate precipitate and the filter and presents little or no safety hazard. The addition of nitric acid during this step is not necessary. Cool the contents of the flask to room temperature.

11.5 Add one mL of a 10 % sodium bromate solution to the flask containing the perchloric acid and transfer the contents of the flask to a 60-mL separatory funnel containing 10 mL of 15 % HDEHP in n-heptane and shake for five minutes. After complete phase separation, draw off the lower aqueous layer and discard. Wash the organic extract twice with 5-mL portions of concentrated perchloric acid for two minutes. Discard the wash solutions.

11.6 Strip the trivalent actinides and lanthanides twice for four minutes each with 10-mL portions of 5M nitric acid containing one mL of 10 % sodium bromate solution. Transfer the solution containing the trivalent actinides and lanthanides to another separatory funnel containing 10 mL of 15 % HDEHP in n-heptane and extract for two minutes to remove any plutonium, thorium, or tetravalent cerium that may have stripped during the back extraction of the trivalent actinides. After complete phase separation, draw off the lower aqueous layer into a 70 mL polycarbonate counting bottle. Any residual

organic will transfer to the sides of the counting bottle.

11.7 Transfer the contents of the counting bottle to a 50-mL conical polycarbonate centrifuge tube. Add 5 mg of lanthanum carrier and 5 mL of concentrated hydrofluoric acid to the centrifuge tube to precipitate the rare earth fluorides. Heat the contents of the centrifuge tube in a hot water bath for ten minutes. Cool the contents to room temperature and centrifuge at 4000 rpm for five minutes. Discard the supernate, add 0.10 mL of concentrated phosphoric acid to the centrifuge tube, and transfer the contents to a TFE fluorocarbon beaker with a small volume of water. Place the beaker on a fiberglass mat-covered hot plate and evaporate the contents to the 0.10 mL of concentrated phosphoric acid previously added.

11.8 Transfer the phosphoric acid to a 50-mL conical polycarbonate centrifuge tube using 3 to 4 mL of water to complete the transfer. Add one drop of a 0.05 % solution of silver nitrate and 3 mL of 0.2M ammonium persulfate solution and adjust the volume to 10 mL. Heat the sample for ten minutes in a hot water bath. Add an additional one mL of ammonium persulfate and continue heating the solution for another two minutes. Add 2 mL of a 0.1M ammonium persulfate-3M ammonium fluoride solution to the centrifuge tube and digest the contents for two minutes in a hot water bath. Remove the centrifuge tube from the hot water bath and cool in an ice-water bath.

11.9 Centrifuge the contents of the centrifuge tube and transfer the supernate containing the oxidized americium to a 50-mL round bottom polycarbonate centrifuge tube. Wash the precipitate with 2-3 mL of the ammonium persulfate-ammonium fluoride solution and add the wash to the supernate. Add one mL of 30 % hydrogen peroxide and heat the supernate in a hot water bath for ten minutes to reduce the hexavalent americium. Remove the round bottom centrifuge tube from the hot water bath, add 50 µg of neodymium carrier, and precipitate the americium fraction using C 1163 Standard Method for Mounting Actinides for Alpha Spectrometry Using Neodymium Fluoride. Submit the sample for alpha spectrometry.

12. Alpha Pulse Height Analysis

12.1 Count the sample on an alpha spectrometer for at least 60 000 s to obtain adequate counting statistics.

12.2 Separately measure the detector background and the reagent blank in the energy range of the americium isotopes and correct the total count of each peak.

13. Calculations

13.1 The concentration of americium-241 in the aliquot of soil taken for analysis is calculated as follows:

$$Am_{241} = \left[\frac{\frac{G_{241}}{t_G} - \frac{B_{241}}{t_B}}{\frac{G_{234}}{t_G} - \frac{B_{243}}{t_B}} \right] \frac{Am_{243}}{W_s}$$

where:

- Am_{241} = concentration of americium-241 in Bq/g,
- G_{241} = the gross counts in the total americium-241 energy region in t_G seconds,
- t_G = length of sample count in seconds,

- B_{241} = reagent blank counts in the total americium–241 energy region in t_B seconds,
 t_B = length of reagent blank counts in seconds,
 G_{243} = gross counts in the total americium–243 energy region in t_G seconds,
 B_{243} = detector background counts in the total americium–243 energy region in t_B seconds,
 Am_{243} = the activity of the added americium–243 in Bq, and
 W_s = the weight of sample in g from step 11.1.

13.2 The standard deviation of the americium–241 result, based on counting statistics, can be determined as follows:

$$S_{241} = |Am_{241}| \cdot \left[\frac{\left(\frac{G_{241}}{t_G^2} + \frac{B_{241}}{t_B^2} \right)}{\left(\frac{G_{241}}{t_G} - \frac{B_{241}}{t_B} \right)^2} + \frac{\left(\frac{G_{243}}{t_G^2} + \frac{B_{243}}{t_B^2} \right)}{\left(\frac{G_{243}}{t_G} - \frac{B_{243}}{t_B} \right)^2} + \frac{S_{243}^2}{Am_{243}^2} + \frac{S_{W_s}^2}{W_s^2} \right]^{1/2}$$

where:

- S_{241} = standard deviation of the americium–241 concentration,
 S_{243} = the standard deviation associated with the standard americium–243 value, and
 S_{W_s} = the standard deviation associated with the sample weight.

The first two terms give the contribution due to counting statistics alone. The third term gives the contribution due to the uncertainty in the standard and the last term the uncertainty due to sample weighing.

13.3 The expression in 13.2 for the uncertainty in the reported value of americium–241 includes only contributions from counting statistics, weighing uncertainty, and uncertainty in the standard. The actual uncertainty may also be affected by sample preparation, position, or matrix effects. It is important to verify the computed uncertainty value with the variation from actual repeated runs. A measurement control program designed to track assays of a control standard will give valuable information on method stability and variability.

14. Precision and Bias

14.1 Precision:

14.1.1 A Synthetic Natural Matrix Standard (SNMS) soil was prepared⁶ using NIST–traceable americium–241 as one radionuclide. The soil was analyzed for americium–241 using this test method. The applicable results are collected in the table and show that the precision of the test method for the 14 measured samples has a relative standard deviation of 6 %.

Summary of Americium-241 Results ^A	
Weight of Sample, g	Americium-241, mBq/g
5.0	28.83
5.0	28.17
7.5	30.00
7.5	27.33
10.0	27.83
10.0	31.51
10.01	27.50
10.06	30.67
10.49	31.33
10.12	31.50
10.70	27.50
11.04	28.33
10.11	27.17
10.20	29.17

^A Number of samples = 14

Mean = 29.06

Standard deviation = 1.66

Standard deviation of the mean = 0.44

Known = 29.70

14.1.2 The NIST SRM–4350B, Columbia River Sediment, may also be useful material to verify this test method.

14.2 Bias:

14.2.1 No statistically significant bias (at the 5 % significance level) was observed in the above analysis of this material.

15. Keywords

15.1 alpha spectrometry; Americium–241; coprecipitation; neodymium fluoride mounting; organic extraction; pyrosulfate fusion

⁶ D. G. Olson and R. P. Bernabee, "Preparation and Analysis of High-Quality Spiked Soil Standards," *Health Physics*, Volume 54, Number 4, 1988, pp. 451–459.

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