Designation: C1475 – 05

Standard Guide for Determination of Neptunium-237 in Soil[1]

This standard is issued under the fixed designation C1475; 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 (ε) indicates an editorial change since the last revision or reapproval.

1. Scope

1.1 This guide covers the determination of neptunium-237 in soil by means of radiochemical separations and alpha spectrometry. This guide provides options in the selection of sample preparation, separation, and measurement. Although neptunium-237 is not a component of global fallout, it is a product of production reactors and spent fuel processing. This guide is designed for analysis of ten grams of soil previously collected and treated in accordance with Practices C998 and C999. Larger-size samples of environmental soil may also be analyzed, as long as the concentrations of interferences such as uranium and thorium are at or near environmental concentrations. Depending on the choice of a sample dissolution method, all chemical forms of neptunium may not be completely solubilized. This guide should allow the determination of neptunium-237 concentrations from sub becquerel per gram levels to applicable standards depending on count time, sample size, detector efficiency, background, and tracer yield.

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.* Specific precautionary statements are given in Section 8.

2. Referenced Documents

2.1 *ASTM Standards:*[2]

C998 Practice for Sampling Surface Soil for Radionuclides
C999 Practice for Soil Sample Preparation for the Determination of Radionuclides
C1000 Test Method for Radiochemical Determination of Uranium Isotopes in Soil by Alpha Spectrometry
C1001 Test Method for Radiochemical Determination of Plutonium in Soil by Alpha Spectroscopy
C1163 Practice for Mounting Actinides for Alpha Spectrometry Using Neodymium Fluoride
C1284 Practice for Electrodeposition of the Actinides for Alpha Spectrometry
C1317 Practice for Dissolution of Silicate or Acid-Resistant Matrix Samples
C1342 Practice for Flux Fusion Sample Dissolution
C1387 Guide for the Determination of Technetium-99 in Soil
C1412 Practice for Microwave Oven Dissolution of Glass Containing Radioactive and Mixed Wastes
D1193 Specification for Reagent Water
D1890 Test Method for Beta Particle Radioactivity of Water
D3084 Practice for Alpha-Particle Spectrometry of Water
D4962 Practice for NaI(Tl) Gamma-Ray Spectrometry of Water
IEEE/ASTM SI-10 Standard for the Use of the International System of Units (SI): The Modern Metric System

3. Summary of Guide

3.1 This guide may be used to determine neptunium-237 in soil at potential release sites. A neptunium-239 yield monitor is added to the soil aliquot and the sample solubilized by one of several methods, such as those described in Guide C1387. The neptunium is separated from the resulting solution using an extraction chromatography column. A valence adjustment is performed prior to loading the sample onto the conditioned chromatography column. The sample is passed through the column, which retains the neptunium in the +4 oxidation state. The column is washed to remove interferences and selectively eluted with dilute acid. The samples are prepared for measurement by neodymium fluoride co-precipitation or electrodeposition, and the neptunium-237 content determined by alpha spectrometry. The neptunium-239 yield monitor is determined by beta or gamma-ray counting.

4. Significance and Use

4.1 A soil sampling and analysis program provides a direct means of determining the concentration and distribution of radionuclides in soil. The presence and extent of neptunium-237 is of particular interest because it is one of the more mobile transuranics in terms of migration and plant uptake. Since soil is an integrator and a reservoir on long-lived radionuclides, and serves as an intermediary in several pathways of potential

---

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

Current edition approved June 1, 2005. Published July 2005. Originally approved in 2000. Last previous edition approved in 2000 as C1475 – 00. DOI: 10.1520/C1475-05.

[2] 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.

importance to humans, knowledge of the concentration of neptunium-237 in soil is essential.

5. Interferences

5.1 Phosphates present in the sample matrix will interfere with the separation chemistry. Aluminum nitrate may be added to the load solution to minimize this problem.

5.2 High concentrations of uranium or thorium can overload the column, resulting in low recoveries of neptunium or spectral interferences. A preliminary separation may be required to avoid spectral interference from uranium, see Test Method C1000. The presence of uranium-233 and uranium-234 could cause spectral interferences with the neptunium-237 determination.

6. Apparatus

6.1 Apparatus for the sample dissolution and measurement are identified within the selected test method.

6.2 *Extraction Columns*, with a bed volume of several millilitres for the extraction chromatography resin.[3]

7. Reagents

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.[4] 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, references to water shall be understood to mean reagent water as defined in Specification D1193.

7.3 *Aluminum Nitrate Hydrate* $[Al(NO_3)_3 \cdot 9H_2O]$.

7.4 *Aliphatic Quaternary Amine Extraction Chromatography Resin.*[3]

7.5 *Ferrous Sulfamate* $[Fe(SO_3NH_2)_2]$*, 1.0 M*—Dissolve 38.8 g of sulfamic acid (NH_2SO_3H) and 11.6 g of iron powder in approximately 150 mL of water. Stir while heating until dissolved and then dilute to 200 mL with water. Prepare fresh each week or keep under a nitrogen blanket to minimize oxidation.

7.6 *Hydrofluoric Acid (48 to 51 %)*—Concentrated hydrofluoric acid (HF).

7.7 *Iron Powder*.

7.8 *Nitric Acid (sp gr 1.42)*—Concentrated nitric acid (HNO_3).

7.9 *Nitric Acid, 3.0 M*—Add 189 mL of concentrated nitric acid to 400 mL of water and dilute to 1.0 L with water and mix.

7.10 *2.5 M Nitric Acid-0.5 M Aluminum Nitrate*—Dissolve 187.6 g of aluminum nitrate in about 500 mL of water, add 159 mL of concentrated nitric acid, and dilute to 1.0 L.

7.11 *0.02 M Nitric Acid-0.02 M Hydrofluoric Acid*—Add 1.25 mL of concentrated nitric acid and 0.7 mL of concentrated hydrofluoric acid to 800 mL of water and dilute to 1.0 L with water and mix.

7.12 *Radiometric Yield Tracer*—Neptunium-239 is normally used as a yield monitor in the determination of neptunium-237. Where beta counting is used to determine the neptunium-239 yield, a relatively low amount of activity, for example, 5 to 15 Bq, is typically required to obtain the desired precision with a 60-min count duration. Another option is the addition of a known quantity of americium-243, in secular equilibrium with neptunium-239, directly to the sample. This approach has the advantage of allowing the use of americium-243 solutions of NIST-traceable activity. Where gamma-ray counting is used to determine the neptunium-239 yield, a larger amount of activity, for example, 120 to 1200 Bq, may be required to obtain the desired precision with a 10-min count duration dependent on the use of a NaI(TI) or HPGe detector. In this situation, it may be preferred to obtain the neptunium-239 from an americium-243 ′cow.′[5] This approach has the advantage of conserving the americium-243 parent with the neptunium-239 activity being replenished over time. However, it has the disadvantage that the neptunium-239 activity of the milked solution must be determined with high precision in order to not adversely impact the precision of the sample neptunium-237 activity determination.

7.13 *Sodium Nitrite* $(NaNO_2)$.

7.14 *Sodium Nitrite, 3.0 M*—Dissolve 2.1 g of sodium nitrite in 10 mL of water. Prepare fresh daily.

7.15 If prepacked columns are not available, prepare the resin by combining it with an equal volume of water, mix well, and allow to settle overnight. Decant the supernatant water. Prepare a column for each sample by pouring resin into each column. Adjust the settled resin bed volume to approximately 2 mL. Perpare only enough resin that is needed for use each day.

7.16 The use of dry-packed cartridges of the extraction chromatography resin in combination with a vacuum manifold system has been found acceptable. When using vacuum-assisted flow, it is essential to ensure that flow rates do not exceed 1 mL/min for load solutions and strip solutions, and 2 mL/min for the rinse solutions. Alternately, follow the manufacturer's recommendations.

8. Precautions

8.1 Refer to the laboratory's chemical hygiene plan and other applicable guidance for handling chemical and radioactive materials and for the management of radioactive, mixed, and hazardous waste.

[3] Prepacked columns of TEVA Resin from Eichrom Technologies, Inc., Darien, IL, have been found to be satisfactory for this purpose.

[4] *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*, BDH Ltd., Poole, Dorset, U.K., and the *United States Pharmacopeia and National Formulary*, U.S. Pharmacopeial Convention, Inc. (USPC), Rockville, MD.

[5] Information on the generation and use of americium-243 cows to obtain neptunium-239 can be obtained from: Garraway, J., and Wilson, P. D., "Preparation of Np-239 by Separation from the Parent Am-243," *Journal of the Less-Common Metals*, 91, 1983, L13-L16, and Bubernak, J., Lew, M. S., and Matlack, G. M., "Ion Exchange Extraction, Separation and Radiochemical Determination of Neptunium-237 in Plutonium-238," *Analytica Chemica Acta*, 48, 1969, pp. 233-241.