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# Standard Guide for Determination of Plutonium and Neptunium in Uranium Hexafluoride and U-Rich Matrix by Alpha Spectrometry<sup>1</sup>

This standard is issued under the fixed designation C1561; 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 method covers the determination of plutonium and neptunium isotopes in uranium hexafluoride by alpha spectroscopy. The method ~~could~~ can also be applicable to any matrix that may be converted to a nitric acid system.

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 requirements prior to use.*

## 2. Referenced Documents

### 2.1 ASTM Standards:<sup>2</sup>

C787 Specification for Uranium Hexafluoride for Enrichment

C996 Specification for Uranium Hexafluoride Enriched to Less Than 5 % <sup>235</sup>U

C1163 Practice for Mounting Actinides for Alpha Spectrometry Using Neodymium Fluoride

C1475 Guide for Determination of Neptunium-237 in Soil

D1193 Specification for Reagent Water

D3084 Practice for Alpha-Particle Spectrometry of Water

D3648 Practices for the Measurement of Radioactivity

## 3. Terminology

3.1 *reagent blank*—DI water processed the same as the samples; used in the determination of the minimum detectable activity.

3.2 *region-of-interest (ROI)*—the channels, or region, in the alpha spectra in which the counts due to a specific radioisotope appear on a functioning calibrated alpha spectrometry system.

## 4. Summary of Test Method

4.1 An aliquot of hydrolyzed uranium hexafluoride equivalent to approximately 0.5 g of uranium is converted to an oxalic acid-nitric acid system and the uranium is selectively removed via solid phase extraction. Plutonium and neptunium are further purified by additional solid phase extractions. The plutonium and neptunium are then co-precipitated with neodymium as the fluorides and counted by alpha spectrometry.

4.2 Tracer recoveries using this method are typically between 75 and 90%. The resolution of the tracer is typically less than 40 keV full-width at half-maximum.

4.2 Tracer recoveries using this method are typically between 75 and 90 % for uranium hexafluoride, for different matrix (with impurities): ~ 10 %. The resolution of the tracer is typically less than 40 keV full-width at half-maximum.

4.3 The minimum detectable activity will vary with tracer recovery, sample size, instrument background, and counting efficiency.

## 5. Significance and Use

5.1 The method is applicable to the analysis of materials to demonstrate compliance with the specifications set forth in Specifications C787 and C996.

5.2 The method can be used to quantify Pu and Np in U-rich matrix before to recycle them.

<sup>1</sup> 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.

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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.

## 6. Interferences

6.1 Incomplete removal of U-234 from the neptunium fraction could result in a false positive for the Np-237 analysis. The method has been shown to adequately remove uranium at enrichments up to 5 %. If the method is used for the analysis of materials at greater than 5 % enrichment, a blank consisting of uranium at the same enrichment as the samples should be analyzed to show adequate removal of the U-234.

6.2 A Pu tracer is used to monitor the chemical recovery of the Np. Spiked analyses should be performed to confirm the appropriateness of this correction; fractionation of Np and Pu during the separation could lead to incorrect test results.

## 7. Instrumentation

7.1 *Alpha Spectrometry System*—See Practices D3084 and D3648 for a description of the apparatus.

## 8. Apparatus

8.1 *Ion Exchange Columns*, able to hold a 10 mL resin bed and 15 mL solution washes.

8.2 *Filter Paper*, 0.1  $\mu\text{m}$  pore size, 25-mm diameter, compatible with HF.<sup>3</sup>

## 9. Reagents and Materials

9.1 *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>

9.2 *Purity of Water*—Unless otherwise indicated, references to water shall be understood to mean reagent water as defined in Specification D1193.

9.3 *Ammonium Oxalate (0.1M)*—Dissolve 12.4 g ammonium oxalate in approximately 500 mL of water and dilute to 1 L.

9.4 *Ascorbic Acid Solution (Saturated)*—Add ascorbic acid to 2M nitric acid while stirring until no more ascorbic acid will dissolve. Prepare fresh when needed for use.

9.5 *Ethanol*, ethyl alcohol, absolute (200 proof), denatured.

9.6 *Hydrochloric Acid (HCl)*, specific gravity 1.19, concentrated.

9.7 *Hydrochloric Acid, 9M*—Add 750 mL concentrated HCl to 100 mL of water, dilute to a final volume of 1 L.

9.8 *Hydrochloric Acid, 4M*—Add 333 mL of concentrated HCl to 500 mL of water; dilute to a final volume of 1 L.

9.9 *Hydrochloric Acid, 1.5M*—Add 125 mL of concentrated HCl to 500 mL of water; dilute to a final volume of 1 L.

9.10 *Hydrochloric Acid, 1M*—Add 83 mL of concentrated HCl to 500 mL of water; dilute to a final volume of 1 L.

9.11 *Hydrofluoric Acid (HF)*, concentrated HF, minimum assay 48 %.

9.12 *Iron (III) Nitrate (10 mg Fe/mL)*—Dissolve 18.0 g of  $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$  in 250 mL of water.

9.13 *Neodymium Chloride (10 mg Nd/mL)*—Add 25 mL concentrated HCl to 1.17 g neodymium oxide and heat at 100°C until dissolved. Allow solution to cool and dilute to 100 mL with water.

9.14 *Neodymium Chloride (100  $\mu\text{g}$  Nd/mL)*—Dilute 1 mL of the 10 mg Nd/mL solution to 100 mL with water.

9.15 *Nitric Acid (HNO<sub>3</sub>)*, concentrated nitric acid, specific gravity 1.42.

9.16 *Nitric Acid (3M)*—Add 188 mL concentrated nitric acid to 500 mL of water; dilute to a final volume of 1 L.

9.17 *Nitric Acid (2M)*—Add 125 mL of concentrated nitric acid to 500 mL of water; dilute to a final volume of 1 L.

9.18 *Oxalic Acid in 1M HCl (0.1M)*—Dissolve 12.6 g oxalic acid dihydrate in 500 mL of 1M HCl; dilute to a final volume of 1 L with 1M HCl.

9.19 *Oxalic Acid in 2M HNO<sub>3</sub> (0.1M)*—Dissolve 12.6 g oxalic acid dihydrate in 500 mL of 2M HNO<sub>3</sub>; dilute to a final volume of 1 L with 2M HNO<sub>3</sub>.

9.20 *Pu-236 or Pu-242 Tracer*, traceable to a national or international standard.

9.21 *Sodium Nitrite (100 mg/mL)*—Dissolve 500 mg NaNO<sub>2</sub> in 5 mL water. Prepare fresh when using.

9.22 *Extraction Chromatography Resin*, containing octylphenyl-N,N-di-isobutyl carbamoylphosphine oxide (CMPO) dissolved in tri-n-butyl phosphate (TBP) as the immobilized extractant.<sup>5,6</sup>

9.23 *Extraction Chromatography Resin*, containing diamyl amylophosphonate (DAAP) as the immobilized extractant.<sup>7,8</sup>

## 10. Calibration and Standardization

10.1 The alpha spectrometry units should be calibrated for energy, resolution and efficiency according to the manufacturers

<sup>3</sup> The Gelman Metrcel filter has been found to be acceptable.

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

<sup>5</sup> Horwitz, E. P., Chiarizia, R., Dietz, M. L., Diamond, H., and Nelson, D., "Separation and Preconcentration of Actinides from Acidic Media by Extraction Chromatography," *Analytica Chimica Acta*, 281, 1993, pp. 361-372.

<sup>6</sup> TRU resin from EIChroM Industries, Darien IL, USA, has been found to be acceptable.

<sup>7</sup> Horwitz, E. P., Dietz, M. L., Chiarizia, R., Diamond, H., Essling, A. M., and Graczyk, D., "Separation and Preconcentration of Uranium from Acidic Media by Extraction Chromatography," *Analytica Chimica Acta*, 266, 1992, pp. 25-37.

<sup>8</sup> U-TEVA resin from EIChroM Industries, Darien IL, USA, has been found to be acceptable.