



Designation: E321 – 20

Standard Test Method for Atom Percent Fission in Uranium and Plutonium Fuel (Neodymium-148 Method)¹

This standard is issued under the fixed designation E321; 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 test method covers the determination of stable fission product ¹⁴⁸Nd in irradiated uranium (U) fuel (with initial plutonium (Pu) content from 0 to 50 %) as a measure of fuel burnup (1-3).²

1.2 It is possible to obtain additional information about the uranium and plutonium concentrations and isotopic abundances on the same sample taken for burnup analysis. If this additional information is desired, it can be obtained by precisely measuring the spike and sample volumes and following the instructions in Test Method E267.

1.3 The values stated in SI units are to be regarded as standard. No other units of measurement are included in this standard.

1.4 *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, health, and environmental practices and determine the applicability of regulatory limitations prior to use.*

1.5 *This international standard was developed in accordance with internationally recognized principles on standardization established in the Decision on Principles for the Development of International Standards, Guides and Recommendations issued by the World Trade Organization Technical Barriers to Trade (TBT) Committee.*

2. Referenced Documents

2.1 *ASTM Standards:*³

C859 Terminology Relating to Nuclear Materials

C1168 Practice for Preparation and Dissolution of Plutonium

Materials for Analysis

C1267 Test Method for Uranium by Iron (II) Reduction in Phosphoric Acid Followed by Chromium (VI) Titration in the Presence of Vanadium

C1347 Practice for Preparation and Dissolution of Uranium Materials for Analysis

C1411 Practice for The Ion Exchange Separation of Uranium and Plutonium Prior to Isotopic Analysis

C1415 Test Method for ²³⁸Pu Isotopic Abundance By Alpha Spectrometry

C1625 Test Method for Uranium and Plutonium Concentrations and Isotopic Abundances by Thermal Ionization Mass Spectrometry

C1672 Test Method for Determination of Uranium or Plutonium Isotopic Composition or Concentration by the Total Evaporation Method Using a Thermal Ionization Mass Spectrometer

C1832 Test Method for Determination of Uranium Isotopic Composition by the Modified Total Evaporation (MTE) Method Using a Thermal Ionization Mass Spectrometer

C1845 Practice for The Separation of Lanthanide Elements from Uranium Matrices Using High Pressure Ion Chromatography (HPIC) for Isotopic Analyses by Inductively Coupled Plasma Mass Spectrometry (ICP-MS)

D1193 Specification for Reagent Water

E180 Practice for Determining the Precision of ASTM Methods for Analysis and Testing of Industrial and Specialty Chemicals (Withdrawn 2009)⁴

E244 Test Method for Atom Percent Fission in Uranium and Plutonium Fuel (Mass Spectrometric Method) (Withdrawn 2001)⁴

E267 Test Method for Uranium and Plutonium Concentrations and Isotopic Abundances

3. Terminology

3.1 Definitions:

3.1.1 For definitions of terms used in this test method but not defined herein, refer to Terminology C859.

¹ This test method 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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² The boldface numbers in parentheses refer to the list of references at the end of this test method.

³ 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.

⁴ The last approved version of this historical standard is referenced on www.astm.org.

TABLE 1 K Factors to Correct ^{148}Nd for ^{147}Nd Thermal Neutron Capture^A

Total Neutron Flux, ϕ (neutrons/cm ² /s)	Total Neutron Exposure, ϕt (neutrons/cm ²)				
	1×10^{20}	3×10^{20}	1×10^{21}	2×10^{21}	3×10^{21}
3×10^{12}	0.9985	0.9985	0.9985	0.9985	0.9985
1×10^{13}	0.9956	0.9952	0.9950	0.9950	0.9950
3×10^{13}	0.9906	0.9870	0.9856	0.9853	0.9852
1×10^{14}	0.9858	0.9716	0.9598	0.9569	0.9559
3×10^{14}	0.9835	0.9592	0.9187	0.9008	0.8941
1×10^{15}	0.9826	0.9526	0.8816	0.8284	0.8006

^A Assuming continuous reactor operation and a 274 ± 91 barn ^{147}Nd effective neutron absorption cross section for a thermal neutron power reactor. This cross section was obtained by adjusting the 440 ± 150 barn ^{147}Nd cross section (7) measured at 20 °C to a Maxwellian spectrum at a neutron temperature of 300 °C.

4. Summary of Test Method

4.1 Fission product neodymium (Nd) is chemically separated from irradiated fuel and determined by isotopic dilution mass spectrometry. Enriched ^{150}Nd is selected as the Nd isotope diluent, and the mass-142 position is used to monitor for natural Nd contamination. The two rare earths immediately adjacent to Nd do not interfere. Interference from other rare earths, such as natural or fission product ^{142}Ce or natural ^{148}Sm and ^{150}Sm is avoided by removing them in the chemical purification (4 and 5).

4.2 After addition of a blended ^{150}Nd , ^{233}U , and ^{242}Pu spike to the sample, the Nd, U, and Pu fractions are separated from each other by ion exchange. Each fraction is further purified for mass analysis. Two alternative separation procedures are provided.

4.3 The gross alpha, beta, and gamma decontamination factors are in excess of 10^3 and are normally limited to that value by traces of ^{242}Cm , ^{147}Pm , and ^{241}Am , respectively (and sometimes ^{106}Ru), none of which interferes in the analysis. The 70 ng ^{148}Nd minimum sample size recommended in the procedure is large enough to exceed by 100-fold a typical natural Nd blank of 0.7 ± 0.7 ng ^{148}Nd (for which a correction is made) without exceeding radiation dose rates of 20 $\mu\text{Sv/h}$ (20 mR/h) at 1 m. Since a constant amount of fission products is taken for each analysis, the radiation dose from each sample is similar for all burnup values and depends principally upon cooling time. Gamma dose rates vary from 200 $\mu\text{Sv/h}$ (20 mR/h) at 1 m for 60-day cooled fuel to 20 $\mu\text{Sv/h}$ (2 mR/h) at 1 m for 1-year cooled fuel. Beta dose rates are an order of magnitude greater, but can be shielded out with a 1/2-in. (12.7 mm) thick plastic sheet. By use of such simple local shielding, dilute solutions of irradiated nuclear fuel dissolver solutions can be analyzed for burnup without an elaborate shielded analytical facility. The decontaminated Nd fraction is mounted on a rhenium (Re) filament for mass analysis. Samples from 20 ng to 20 μg run well in the mass spectrometer with both NdO^+ and Nd^+ ion beams present. The metal ion is enhanced by deposition of carbonaceous material on the filament as oxygen getter. (Double and triple filament designs do not require an oxygen getter.)

5. Significance and Use

5.1 The burnup of an irradiated nuclear fuel can be determined from the amount of a fission product formed during irradiation. Among the fission products, ^{148}Nd has the following properties to recommend it as an ideal burnup indicator:

5.1.1 It is not volatile, does not migrate in solid fuels below their recrystallization temperature, and has no volatile precursors.

5.1.2 It is nonradioactive and requires no decay corrections.

5.1.3 It has a low destruction cross section and formation from adjacent mass chains can be corrected for.

5.1.4 It has good emission characteristics for mass analysis.

5.1.5 Its fission yield is nearly the same for ^{235}U and ^{239}Pu and is essentially independent of neutron energy (6).

5.1.6 It has a shielded isotope, ^{142}Nd , which can be used for correcting natural Nd contamination.

5.1.7 It is not a normal constituent of unirradiated fuel.

5.2 The analysis of ^{148}Nd in irradiated fuel does not depend on the availability of preirradiation sample data or irradiation history. Atom percent fission is directly proportional to the ^{148}Nd -to-fuel ratio in irradiated fuel. However, the production of ^{148}Nd from ^{147}Nd by neutron capture will introduce a systematic error whose contribution must be corrected for. In power reactor fuels, this correction is relatively small. In test reactor irradiations where fluxes can be very high, this correction can be substantial (see Table 1).

5.3 The test method can be applied directly to U fuel containing less than 0.5 % initial Pu with 1 to 100 GW days/metric ton burnup. For fuel containing 5 to 50 % initial Pu, increase the Pu content by a factor of 10 to 100, respectively in both reagents 6.3 and 6.4.

6. Reagents and Materials

NOTE 1—For chemical processing (dissolution and separations) use of the appropriate referenced practices in Section 2 is acceptable: Practices C1168, C1347, C1411, and C1845.

6.1 *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.⁵ 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.

⁵ *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.

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

6.3 *Blended ^{148}Nd , ^{239}Pu , and ^{238}U Calibration Standard*—Prepare a solution containing about 0.0400 mg ^{148}Nd /litre, 50 mg ^{238}U /litre, and 2.5 mg ^{239}Pu /litre, in nitric acid (HNO_3 , 1 + 1) with 0.01 M hydrofluoric acid (HF) as follows. With a new calibrated, clean, Kirk-type micropipet, add 0.500 mL of ^{239}Pu known solution (see 6.11) to a calibrated 1-litre volumetric flask. Rinse the micropipet into the flask three times with HNO_3 (1 + 1). In a similar manner, add 0.500 mL of ^{238}U known solution (see 6.12) and 1.000 mL of ^{148}Nd known solution (see 6.9). Add ten drops of concentrated HF and dilute exactly to the 1-litre mark with HNO_3 (1 + 1) and mix thoroughly.

6.3.1 From K_{148} (see 6.9), calculate the atoms of ^{148}Nd /mL of calibration standard, C_{148} , as follows:

$$C_{148} = \frac{\text{mL } ^{148}\text{Nd known solution}}{1000 \text{ mL calibration standard}} \times K_{148} \quad (1)$$

6.3.2 From K_{238} (see 6.12), calculate the atoms of ^{238}U /mL of calibration standard, C_{238} , as follows:

$$C_{238} = \frac{\text{mL } ^{238}\text{U known solution}}{1000 \text{ mL calibration standard}} \times K_{238} \quad (2)$$

6.3.3 From K_{239} (see 6.11), calculate the atoms of ^{239}Pu /mL of calibration standard, C_{239} , as follows:

$$C_{239} = \frac{\text{mL } ^{239}\text{Pu known solution}}{1000 \text{ mL calibration standard}} \times K_{239} \quad (3)$$

6.3.4 Flame seal 3 to 5 mL portions in glass ampoules to prevent evaporation after preparation until time of use. For use, break off the tip of an ampoule, pipet promptly the amount required, and discard any unused solution. If more convenient, calibration solution can be flame-sealed in pre-measured 1000 μL portions for quantitative transfer when needed.

6.4 *Blended ^{150}Nd , ^{233}U , and ^{242}Pu Spike Solution*—Prepare a solution containing about 0.4 mg ^{150}Nd /litre, 50 mg ^{233}U /litre, and 2.5 mg ^{242}Pu /litre in HNO_3 (1 + 1) with 0.01 M HF. These isotopes are obtained in greater than 95, 99, and 99 % isotopic purity, respectively, from the DOE Isotope Business Office located at Oak Ridge National Laboratory.” Standardize the spike solution as follows:

6.4.1 In a 5 mL beaker, place about 0.1 mL of ferrous solution, exactly 500 μL of calibration standard (see 6.3) and exactly 500 μL of spike solution (see 6.4). In a second beaker, place about 0.1 mL of ferrous solution and 1 mL of calibration standard without any spike. In a third beaker, place about 0.1 mL of ferrous solution and 1 mL of spike solution without standard. Mix well and allow to stand for 5 min to reduce Pu (VI) to Pu (III) or Pu (IV).

6.4.2 Follow the procedure described in 9.2.4 – 9.5.8 or 9.6.2 – 9.7.11. Measure the Pu, U, and Nd isotopes by surface ionization mass spectrometry following the procedure described in 9.8.1 – 9.8.3.2. On the Pu fractions, record the atom ratios of ^{242}Pu to ^{239}Pu in the calibration standard, $C_{2/9}$; in the spike solution, $S_{2/9}$; and in the standard-plus-spike mixture, $M_{2/9}$. On the U fractions record the corresponding ^{233}U -to- ^{238}U

ratios, $C_{3/8}$, $S_{3/8}$, and $M_{3/8}$. On the Nd fractions, record the corresponding ^{150}Nd -to- ^{148}Nd ratios, $C_{50/48}$, $S_{50/48}$, and $M_{50/48}$. Correct all average measured ratios for mass discrimination bias (see 8.2).

6.4.3 Calculate the number of atoms of ^{150}Nd /mL of Spike, A_{50} , as follows:

$$A_{50} = C_{148}[(M_{50/48} - C_{50/48})/(1 - M_{50/48}/S_{50/48})] \quad (4)$$

6.4.4 Calculate the number of atoms of ^{233}U /mL of spike, A_{33} , as follows:

$$A_{33} = C_{238}[(M_{3/8} - C_{3/8})/(1 - M_{3/8}/S_{3/8})] \quad (5)$$

6.4.5 Calculate the number of atoms of ^{242}Pu /mL spike, A_{42} , as follows:

$$A_{42} = C_{239}[(M_{2/9} - C_{2/9})/(1 - M_{2/9}/S_{2/9})] \quad (6)$$

6.4.6 Store in the same manner as the calibration standard (see 6.3), that is, flame seal 3 to 5 mL portions in glass ampoules. For use, break off the tip of an ampoule, pipet promptly the amount required, and discard any unused solution. If more convenient, spike solution can be flame sealed in a premeasured 1000 μL portions for quantitative transfer to individual samples.

6.5 *Ferrous Solution (0.001 M)*—Add 40 mg of reagent grade ferrous ammonium sulfate ($\text{Fe}(\text{NH}_4)_2(\text{SO}_4)_2 \cdot 6\text{H}_2\text{O}$) and 1 drop of concentrated H_2SO_4 to 5 mL of redistilled water. Dilute to 100 mL with water and mix. This solution does not keep well. Prepare fresh daily.

6.6 *Filament Mounting Solution*—Dissolve 70 mg of sucrose in 100 mL of water (single filament only).

6.7 *Hydrofluoric Acid*—Reagent grade concentrated HF (28 M).

6.8 *Methanol*, absolute.

6.9 *^{148}Nd Known Solution*—Heat natural Nd_2O_3 (>99.9 % pure) in an open crucible at 900 °C for 1 h to destroy any carbonates present and cool in a dessicator. Weigh 0.4071 g of Nd_2O_3 and place it in a calibrated 500 mL volumetric flask. Dissolve the oxide in HNO_3 (1 + 1) and dilute to the 500 mL mark with HNO_3 (1 + 1) and mix thoroughly. By using the weight of Nd_2O_3 in grams, and the purity, calculate the atoms of ^{148}Nd /mL of known solution, K_{148} , as follows:

$$K_{148} = \text{g Nd}_2\text{O}_3 / 500 \text{ mL} \times \% \text{ purity} / 100 \quad (7)$$

$$\times 50.38 \text{ mg } ^{148}\text{Nd} / 1 \text{ g Nd}_2\text{O}_3 \times (6.025$$

$$\times 10^{20} \text{ atoms}) / 147.92 \text{ molecular weight}$$

6.10 *Perchloric Acid*—70 % HClO_4 .

6.11 *^{239}Pu Known Solution*—Add 10 mL of HCl (1 + 1) to a clean calibrated 100 mL flask. Cool the flask in an ice water bath. Allow time for the acid to reach approximately 0 °C and place the flask in a glove box. Displace the air in the flask with inert gas (Ar, He, or N_2). Within the glove box, open the U.S. National Institute of Standards and Technology Plutonium Metal Standard Sample 949, containing about 0.5 g of Pu (actual weight individually certified), and add the metal to the cooled HCl. After dissolution of the metal, dilute to the 100 mL mark with HCl.