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 ^{148}Nd 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 *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.*

2. Referenced Documents

2.1 *ASTM Standards:*[3]

D1193 Specification for Reagent Water

E180 Practice for Determining the Precision of ASTM Methods for Analysis and Testing of Industrial and Specialty Chemicals[4]

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

E267 Test Method for Uranium and Plutonium Concentrations and Isotopic Abundances

3. Summary of Test Method

3.1 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).

3.2 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.

3.3 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 μ 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 μ Sv/h (20 mR/h) at 1 m for 60-day cooled fuel to 20 μ 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 ½-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.)

4. Significance and Use

4.1 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: (*1*) It is not volatile, does not migrate in solid fuels below their recrystallization temperature, and has no volatile precursors. (*2*) It is nonradioactive and requires no decay corrections. (*3*) It has a low destruction cross section and formation from adjacent mass chains can be corrected for. (*4*) It has good emission characteristics for mass analysis. (*5*) Its fission yield is nearly the same for ^{235}U and ^{239}Pu and is essentially independent of neutron energy **(6)**. (*6*) It has a shielded isotope, $_{142}$Nd, which can be used for correcting natural Nd contamination. (*7*) It is not a normal constituent of unirradiated fuel.

4.2 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).

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

5.3 *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 5.11) 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 5.12) and 1.000 mL of ^{148}Nd known solution (see 5.9). Add 10 drops of concentrated HF and dilute exactly to the 1-litre mark with HNO$_3$ (1 + 1) and mix thoroughly.

5.3.1 From $K_{1\,48}$ (see 5.9), calculate the atoms of ^{148}Nd/mL of calibration standard, C_{148}, as follows:

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

5.3.2 From K_{238} (see 5.12), calculate the atoms of ^{238}U/mL of calibration standard, C_{238}, as follows:

$$C_{23\,8} = \frac{\text{mL } ^{238}\text{U known solution}}{1000 \text{ mL calibration standard}} \times K_{238} \qquad (2)$$

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

Total Neutron Flux, ϕ (neutrons/cm²/s)	Total Neutron Exposure, ϕI (neutrons/cm²)				
	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.

4.3 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 5.3 and 5.4.

5. Reagents and Materials

5.1 *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.

5.3.3 From K_{239} (see 5.11), calculate the atoms of ^{239}Pu/mL of calibration standard, $C_{23\,9}$, as follows:

$$C_{2\,39} = \frac{\text{mL } ^{239}\text{Pu known solution}}{1000 \text{ mL calibration standard}} \times K_{239} \qquad (3)$$

5.3.4 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.

5.4 *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 Isotopes Sales Department of Oak Ridge National Laboratory. Standardize the spike solution as follows:

5.4.1 In a 5-mL beaker, place about 0.1 mL of ferrous solution, exactly 500 µL of calibration standard (see 5.3) and exactly 500 µL of spike solution (see 5.4). In a second beaker, place about 0.1 mL of ferrous solution and 1 mL of calibration

---

[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.

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).

5.4.2 Follow the procedure described in 7.2.4-7.5.8 or 7.6.2-7.7.11. Measure the Pu, U, and Nd isotopes by surface ionization mass spectrometry following the procedure described in 7.8.1-7.8.3.2 . 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 6.2).

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

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

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

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

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

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

5.4.6 Store in the same manner as the calibration standard (see 5.3), 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.

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

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

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

5.8 *Methanol*, absolute.

5.9 ^{148}*Nd Known Solution*—Heat natural Nd$_2$O$_3$ (>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$_2$O$_3$ 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$_2$O$_3$ in grams, and the purity, calculate the atoms of ^{148}Nd/mL of known solution, K_{148}, as follows:

$$K_{148} = g\ Nd_2O_3/500\ mL \times \%\ purity/100$$
$$\times 50.38mg\ ^{148}Nd/1\ g\ Nd_2O_3 \times (6.025$$
$$\times 10^{20}\ atoms)/147.92\ molecular\ weight \quad (7)$$

5.10 *Perchloric Acid*—70 % HClO$_4$.

5.11 ^{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 is complete, add 1 drop of concentrated HF and 40 mL of HNO$_3$ (1 + 1) and swirl. Place the flask in a stainless-steel beaker for protection and invert a 50-mL beaker over the top and let it stand for at least 8 days to allow any gaseous oxidation products to escape. Dilute to the mark with HNO$_3$ (1 + 1) and mix thoroughly. By using the individual weight of Pu in grams, the purity, and the molecular weight of the Pu given on the NIST certificate, with the atom fraction, A_9, determined as in 8.8, calculate the atoms of ^{239}Pu/mL of ^{239}Pu known solution, K_{239}, as follows:

$$K_{239} = [(mg\ Pu/100\ mL\ solution) \times \%\ purity/100$$
$$\times (6.025 \times 10^{20}\ atoms/Pu\ molecular\ weight) \times A_9] \quad (8)$$

5.12 ^{238}*U Known Solution*—Heat U$_3$O$_8$ from the National Institute of Standards and Technology Natural Uranium Oxide Standard Sample 950 in an open crucible at 900°C for 1 h and cool in a dessicator in accordance with the certificate accompanying the standard sample. Weigh about 12.0 g of U$_3$O$_8$ accurately to 0.1 mg and place it in a calibrated 100-mL volumetric flask. Dissolve the oxide in HNO$_3$ (1 + 1). Dilute to the 100-mL mark with HNO$_3$ (1 + 1) and mix thoroughly. By using the measured weight of U$_3$O$_8$ in grams, the purity given on the NIST certificate, and the atom fraction ^{238}U, A_8, determined as in 8.5, calculate the atoms ^{238}U/mL of ^{238}U solution, K_{238}, as follows:

$$K_{238} = [(g\ U_3O_8/100\ mL\ solution) \times (\%\ purity/100$$
$$\times 848.0\ mg\ U/1\ g\ U_3O_8) \times (6.025$$
$$\times 10^{20}\ atoms/238.03\ molecular\ weight) \times A_8] \quad (9)$$

5.13 *Reagents and Materials for Procedure A*:

5.13.1 *Dowex AGMP-1 Resin*—Convert Dowex AGMP-1 (200 to 400 mesh) chloride form resin[6] to nitrate form by washing 200 mL of resin in a suitable column (for example, a 250-mL buret) with HNO$_3$ (1 + 1) until a drop of effluent falling into an AgNO$_3$ solution remains clear. Finally, rinse with water, and dry overnight in a vacuum dessicator. Store the resin in an airtight container. Since the elution characteristics of ion exchange resins depend upon their actual percentage cross linkage and particle size (surface-to-volume ratio), which may vary from one lot to the next, it is most convenient to set aside a bottle of resin to be used solely for this procedure. Before use on actual samples, a small amount of tracer ^{147}Nd should be taken through the procedure. Collect each consecutive 80 mm fraction of eluant and count for γ radioactivity. If over 80 % of the ^{147}Nd appears in the Nd fraction, the resin can be used as directed; if not, small adjustments can be made in the elution volumes collected.

5.13.2 *Hydrochloric Acid*[7]—Prepare reagent low in U and dissolved solids by saturating redistilled water in a polyethylene container to 12 *M* with HCl gas which has passed through

---

[6] Dowex resin (AGMP-1 or AG1-X4, 200–400 mesh) obtained from Bio-Rad Laboratories, 3300 Regatta Blvd., Richmond, CA, has been found satisfactory.

[7] Ultrex and Ultrex II, or equivalent, ultrapure reagent obtained from J. T. Baker Chemical Co., 222 Red School Lane, Phillipsburg, NJ, has been found satisfactory.