

Standard Practice for Analysis of Spent Nuclear Fuel to Determine Selected Isotopes and Estimate Fuel Burnup¹

This standard is issued under the fixed designation C1769; 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 A sample of spent nuclear fuel is analyzed to determine the quantity and atomic ratios of uranium and plutonium isotopes, neodymium isotopes, and selected gamma-emitting nuclides (¹³⁷Cs, ¹³⁴Cs, ¹⁵⁴Eu, ¹⁰⁶Ru, and ²⁴¹Am). Fuel burnup is calculated from the ¹⁴⁸Nd-to-fuel ratio as described in this method, which uses an effective ¹⁴⁸Nd fission yield calculated from the fission yields of ¹⁴⁸Nd for each of the fissioning isotopes weighted according to their contribution to fission as obtained from this method. The burnup value determined in this way requires that values be assumed for certain reactordependent properties called for in the calculations (**1**, **2**).²

1.2 Error associated with the calculated burnup values is discussed in the context of contributions from random and potential systematic error sources associated with the measurements and from uncertainty in the assumed reactor-dependent variables. Uncertainties from the needed assumptions are shown to be larger than uncertainties from the isotopic measurements, with the largest effect arising from the value of the fast fission factor. Using this factor will provide the most consistent burnup value between calculated changes in heavy element isotopic composition.

1.3 This standard practice contains explanatory notes that are not part of the mandatory portion of the standard.

1.4 The values stated in SI units are to be regarded as the standard. Mathematical equivalents are given in parentheses.

1.5 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
- C1625 Test Method for Uranium and Plutonium Concentrations and Isotopic Abundances by Thermal Ionization Mass Spectrometry
- C859 Terminology Relating to Nuclear Materials
- D1193 Specification for Reagent Water
- E244 Test Method for Atom Percent Fission in Uranium and Plutonium Fuel (Mass Spectrometric Method) (Withdrawn 2001)⁴

3. Terminology

3.1 *Definitions*—For definitions of other standard terms in this practice, refer to Terminology C859.

3.2 Definitions of Terms Specific to This Standard:

3.2.1 gigawatt days per metric ton—the gigawatt days of heat produced per metric ton of uranium plus plutonium initially present in a nuclear fuel.

3.2.2 *heavy element atom percent fission*—the number of fissions per 100 uranium plus plutonium atoms initially present in a nuclear fuel.

3.3 *Symbols:* Symbols used in the procedural equations are defined as follows:

3.3.1 F_5 , F_9 , F_1 , F_8 —heavy element atom percent fission from fission ²³⁵U, ²³⁹Pu, ²⁴¹Pu, and ²³⁸U.

3.3.2 F_T —total heavy element atom percent fission.

3.3.3 F_8^0 , N_5^0 —heavy element atom percent ²³⁸U and ²³⁵U, in the pre-irradiated fuel.

3.3.4 $R_{5/8}^{0}$, $R_{5/8}^{0}$, $R_{5/8}^{0}$, $R_{5/8}^{0}$ —atoms ratios of ²³⁵U to ²³⁸U, ²³⁶U to ²³⁵U in the pre-irradiated fuel.

3.3.5 $R_{3/8}$, $R_{3/8}$, $R_{3/8}$ —atom ratios of 235 U to 238 U, 236 U to 238 U, and 236 U to 235 U in the final irradiated sample.

3.3.6 $R_{\%}$, $R_{\%}$, $R_{\%}$ – atom ratios of ²³⁹Pu, ²⁴⁰Pu, ²⁴¹Pu, ²⁴²Pu and to ²³⁸U in the final irradiated sample.

¹ This practice 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, 2015. Published July 2015. DOI: 10.1520/ C1769-15.

 $^{^{2}\,\}mathrm{The}$ boldface numbers in parentheses refer to a list of references at the end of this standard.

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.

 $^{^{\}rm 4}\,{\rm The}$ last approved version of this historical standard is referenced on www.astm.org.

3.3.7 R'_{4} —atom ratio of ²⁴¹Pu to ²³⁸U in the final irradiated sample corrected for neutron capture, fission, and decay during and after irradiation.

3.3.8 v₈–2.67 \pm 0.30 neutrons per fission of ²³⁸U (3).

3.3.9 v₅-2.426 \pm 0.006 neutrons per fission of ²³⁵U (4).

3.3.10 v₀/v₅—ratio of number of neutrons per fission of ²³⁹Pu to ²³⁵U = 1.192 \pm 0.005 (4).

3.3.11 v₁/v₅—ratio of number of neutrons per fission of ²⁴¹Pu to ²³⁵U = 1.237 \pm 0.017 (4).

3.3.12 t'—elapsed time from the end of irradiation to measurement.

3.3.13 t—irradiation time, s.

3.3.14 λI —decay constant of 153 × 10⁻⁹ s⁻¹.

3.3.15 *c*—ratio of the 238 U fission rate of the fission rate from all other sources expressed as equivalent 235 U fission rate.

3.3.16 ϵ —fast fission factor (defined in Ref (5)) which is 1.00 for fully enriched reactors. Typically, ϵ ranges from 1.03 to 1.07 for low enrichment systems.

3.3.17 a_5 —effective ratio of ²³⁵U (*n*, γ) capture-to-fission cross sections obtained from reactor designer, experiment, or machine calculation. If not otherwise available, it may be estimated from Fig. 1 for well-moderated thermal reactors.

3.3.18 a_9 —effective ratio of ²³⁹Pu (n, γ) capture-to-fission cross sections obtained from reactor designer, experiment, or machine calculation. If not otherwise available, it may be estimated from Fig. 2 for well-moderated thermal reactors.

3.3.19 a_1 —effective ratio of ²⁴¹Pu (n, γ) capture-to-fission cross sections = 0.40 \pm 0.15 for thermal reactors Ref (6). Its neutron spectrum dependence has not been measured.

3.3.20 a_8 —effective ratio of ²³⁸U (*n*, γ) capture-to-fission cross sections averaged over a fission spectrum = 0.58 ± 0.45 (3).

3.3.21 r—epithermal index which is a measure of the proportion of epithermal neutrons in a reactor spectrum. In Ref



FIG. 1 Calculated Dependence of a_5 on Neutron Temperature and Epithermal Index, *r*, for Well-Moderated Thermal Reactors



FIG. 2 Calculated Dependence of *a*₉ on Neutron Temperature and Epithermal Index, *r*, for Well-Moderated Thermal Reactors

(7), r is defined and related mathematically to the cadmium ratio. Note that for r = 0 the spectrum is pure Maxwellian.

3.3.22 Φ —neutron flux, neutrons/cm²-s.

3.3.23 σ_I , σ_5 , σ_6 —total neutron absorption cross sections of ²⁴¹Pu, ²³⁵U, and ²³⁶U. For boiling water reactors, typical core average values are 188 × 10⁻²³ cm², 64.6 × 10⁻²³ cm², and 5 × 10⁻²³ cm², respectively. For pressurized water reactors, typical core average values are 155 × 10⁻²³, 55.6 × 10⁻²³ cm², and 8.4 × 10⁻²³ cm², respectively.

3.3.24 *P*—total ²³⁹Pu neutron captures per initial ²³⁸U atom.

4. Summary of Practice

4.1 Atomic ratios of the isotopes ²³⁴U, ²³⁵U, ²³⁶U, to ²³⁸U and ²⁴⁰Pu, ²⁴¹Pu, and ²⁴²Pu to ²³⁹Pu are measured by mass spectrometry in accordance with Test Method C1625 or a similar methodology. The atom percent fission attributed to fission of ²³⁵U, ²³⁸U, ²³⁹Pu, and ²⁴¹Pu are separately calculated and then summed to obtain the total heavy element atom percent fission (**6**, **8**).

4.2 Fission product neodymium (Nd) is chemically separated from irradiated fuel and determined by isotopic dilution mass spectrometry. Enriched ¹⁵⁰Nd is selected as the neodymium isotope diluent and the mass-142 position is used to monitor for natural neodymium contamination. The two rare earths immediately adjacent to neodymium do not interfere. Interference from other rare earths, such as natural or fission product ¹⁴²Ce or natural ¹⁴⁸Sm and ¹⁵⁰Sm is avoided by removing them in the chemical purification (9, 10).

4.3 After addition of a blended ¹⁵⁰Nd, ²³³U, and ²⁴²Pu spike to the sample, the neodymium, uranium, and plutonium fractions are separated from each other by ion exchange. Each fraction is further purified for isotope dilution mass spectrometry analysis. Two alternative separation procedures are provided.

4.4 The gross alpha beta, and gamma decontamination factors are in excess of 10³ and are normally limited to that value by traces of ²⁴²Cm, ¹⁴⁷Pm, and ²⁴¹Am, respectively (and

sometimes ¹⁰⁶Ru), and are insignificant to the analysis. The 70 ng ¹⁴⁸Nd minimum sample size recommended in the procedure is large enough to exceed by 100-fold a typical natural neodymium blank of 0.70 \pm 0.7 ng ¹⁴⁸Nd (for which a correction is made) without exceeding radiation dose rates of 20μ Sv/h (20 mrem/h) at 1 m for 60-day cooled fuel to 20μ Sv/h (2 mrem/h) at 1 m for 1-year cooled fuel. Beta dose rates are an order of magnitude greater, and may be significantly shielded with a 12.7 mm (1/2-in.) 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 neodymium fraction is mounted on a rhenium (Re) filament for isotope dilution mass spectrometry analysis. Samples from 20 ng to 20 µg run well in the mass spectrometer with both NdO⁺ and Nd⁺ ion beams present.

5. Significance and Use

5.1 This standard practice defines a measure of heavy element atom percent fission from which the output of heat during irradiation can be estimated.

5.2 This standard practice is restricted in use to samples where accurate pre-irradiation U and Pu isotopic analysis is available. This data should be available from the fuel manufacture.

5.3 The contribution of 238 U fast fission is not subject to measurement from isotopic analysis. For reactors in which the majority of fissions are caused by thermal neutrons, the contribution may be estimated from the fast fission factors, ε , found in each reactor design document.

5.4 In post-irradiation isotopic analysis, take extreme care to avoid environmental uranium contamination of the sample. This is simplified by using sample sizes in which the amount of each uranium isotope is more than 1000 times the levels observed in a blank carried through the complete chemistry and mass spectrometry procedure employed.

5.5 Take care to make sure that both the pre-irradiation and the post-irradiation samples analyzed are representative. In the pre-irradiation fuel, the ²³⁵U and ²³⁶U atom ratio content may vary from lot to lot. ²³⁶U is not found in naturally uranium in measurable quantity (<2 ppm of a u basis) but forms during irradiation and increases with each successive pass through the fuel cycle. In the post-irradiation normally varies radially and axially. Radial and axial profiles of atom percent fission can be determined by analyzing samples obtained from along the radius or axis of the fuel element. An average value of atom percent fission can be obtained by totally dissolving the fuel to be averaged, and then mixing and analyzing an aliquot of the resultant solution.

5.6 The burnup of an irradiated nuclear fuel can be determined from the amount of a fission product formed during irradiation. Among the fission products, ¹⁴⁸Nd has the following properties to recommend it as an ideal burnup indicator: (1) It is not volatile. (2) It does not migrate in solid fuels below their recrystallization temperature. (3) It has no volatile precursors. (4) It is nonradioactive and requires no decay corrections. (5) It has a low destruction cross section. (6) Formation of ¹⁴⁸Nd from adjacent mass chains can be corrected for. (7) It has adequate emission characteristics for mass analysis. (8) Its fission yield is nearly equivalent for ²³⁵U and ²³⁹Pu. (9) Its fission yield is essentially independent of neutron energy (11). (*10*) It has a shielded isotope, ¹⁴²Nd, which can be used for correcting natural neodymium contamination. (*11*) It is an atypical constituent of unirradiated fuel.

6. Apparatus

- 6.1 Dissolution bomb.⁵
- 6.2 Oven-convection.

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 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, references to water shall be understood to mean reagent grade as defined by Type I of Specification D1193 or water exceeding these specifications.

7.3 Hydrochloric Acid (sp gr 1.19)-concentrated HCl.

7.4 Nitric Acid (sp gr 1.42)-concentrated (HNO₃).

7.5 Hydrobromic Acid (sp gr 1.18)-concentrated (HBr).

7.6 Perchloric Acid (sp gr 1.67)-concentrated.

7.7 Nitric Acid (2 M)-Add 126 mL concentrated nitric acid to a volume of water and dilute, with water, to a final volume to 1000 mL.

7.8 Nitric Acid (0.3 M)-Add 19 mL concentrated nitric acid to a volume of water and dilute, with water, to a final volume to 1000 mL.

7.9 Nitric Acid (0.1 M)-Add 6 mL concentrated nitric acid to a volume of water and dilute, with water, to a final volume to 1000 mL.

7.10 Methanol.

7.11 Nitric Acid/methanol – $(1.56 \text{ M HNO}_3/80 \% \text{ CH}_3\text{OH})$ Add 99 mL concentrated nitric acid to 50 mL of water then 800 mL of methanol and dilute to a final volume of 1000 mL volume with water.

7.12 Dilute nitric acid/methanol – $(0.156 \text{ M HNO}_3/80 \% \text{ CH}_3\text{OH})$ Take a 100 mL aliquot of the solution made in 7.11 and dilute with 800 mL of methanol and dilute to a final volume of 1000 mL.

7.13 Nitric Acid/Methanol (0.10 M HNO₃/80 % CH₃OH) Add 6.3 mL concentrated nitric acid, 800 mL methanol and dilute, with water, to a final volume to 1000 mL volume with water.

⁵ Parr dissolution bomb Model 4749 has shown to be adequate.

7.14 Hydrofluoric Acid (HF, sp gr 1.18) concentrated.

7.15 Hydrochloric/Hydrofluoric Acid (0.05 M HCl/0.001 HF) Add 4.2 mL concentrated HCl to 50 mL of water; separately add 0.04 mL concentrated HF of 50 mL of water. Combine the two diluted acids and dilute to a final volume of 1000 mL.

7.16 Ion Exchange Resins—Anion, quatinary amine (100-200 and 200-400 mesh).

7.17 Exchange resin⁶, transuranic, octylphenyl-N,N-diisobutyl carbamoylphosphine oxide dissolved in tri-n-butyl phosphate (TBP) and placed on a solid support. **Warning**— Hydrofluoric acid is a highly corrosive acid that can severely burn skin, eyes, and mucous membranes. Hydrofluoric acid differs from other acids because the fluoride ion readily penetrates the skin causing destruction of deep tissue layers. Unlike other acids that are rapidly neutralized hydrofluoric acid reactions with tissue may continue for days if left untreated. Familiarization and compliance with the Safety Data Sheet is essential.

8. Operational Procedure

8.1 Inside Hot Cell:

8.1.1 Measure the mass of a portion of the pulverized fuel into the polytetrafluoroethylene (PTFE) cup of a ParrTM bomb. Test samples are typically 50 g originating from a known location in the fuel element.

8.1.2 Add a recently prepared solution of 10 mL of concentrated HNO₃ / 0.3 mL of concentrated HCl / 0.02 mL of concentrated HF into the PTFE insert for the ParrTM bomb.

8.1.3 Seal the $Parr^{TM}$ bomb and place it into a 175°C oven for at least 72 hours.

8.1.4 Transfer, quantitatively, the dissolved sample solution to a tared polyethylene bottle, using 2 M HNO_3 to rinse the PTFE cup.

8.1.5 Add a smal (~0.1 to 0.2 g) amount of the dissolve sample solution into a tared 30 mL polyethylene bottle and dilute to the desired concentration with 0.3 M HNO_3 in preparation for gamma spectroscopy.

8.1.6 Measure the mass of one aliquot of the dissolved sample solution into a beaker containing 1 to 2 drops of $HClO_4$, and evaporate the solution to dryness. The plutonium isotope ratios will be measured with this aliquot.

8.1.7 Measure the mass of a second aliquot of the dissolved sample solution into beakers containing 1 to 2 drops of HClO₄ and a known amount of spike (242 Pu, 233 U, and 150 Nd) solution. Evaporate this solution to dryness.

8.1.8 Place anion exchange resin (100 to 200 mesh) in a column such that the resin bed is 1 cm by 8 cm. Precondition the resin with \sim 10 bed volumes of concentrated HCl.

8.1.9 Dissolve the dried residues in the two beakers in concentrated HCl (\sim 2 mL) and transfer the solutions into their respective preconditioned 1 cm by 8 cm 0.149 to 0.074 mm

(100 to 200 mesh) anion exchange column. Complete the quantitative transfer using small portions of concentrated HCl.

8.1.10 Wash each column with approximately 15 mL of concentrated HCl and collect the fission product eluent in beakers.

8.1.11 Set aside the columns containing the purified uranium and plutonium.

8.1.12 Evaporate the fission product eluent solutions to dryness.

8.1.13 Dissolve the eluent residues in approximately 2 mL of 2 M HNO_3 and again evaporate to dryness to convert these fission product residues to nitrate salts.

8.1.14 Prepare a column containing 2 mL of the transuranic exchange resin (7.17) or use a prepacked column. Condition the columns with 10 bed volumes of 2 M HNO₃.

8.1.15 Dissolve again the fission product residues in 2 M HNO₃ and transfer quantitatively the solutions unto the column.

8.1.16 Wash the columns with approximately 15 mL of 2 M HNO₃ and collect these eluent solutions in beakers.

8.1.17 Set aside the column containing the rare earths elements along with americium.

8.1.18 The resulting samples (consisting of 2 anion exchange and 2 transuranic exchange columns) should be removed from the hot cell and the eluent solutions should be disposed of according to established facility procedures.

8.2 Outside Hot Cell:

8.2.1 Wash each anion-exchange column (containing the uranium and plutonium) with approximately 15 mL of concentrated HBr and collect the plutonium factions in beakers.

8.2.2 Strip the uranium from the columns using approximately 15 mL of 0.05 M HCl / 0.001 M HF (7.14) and collect the uranium fractions in beakers.

8.2.3 Add concentrated HNO_3 to each of the plutonium and uranium eluent solutions and evaporate dryness. Repeat the evaporation in concentrated HNO_3 as needed to ensure removal of the bromine, as evidenced by a lack of red color in the dried residue.

8.2.4 Strip the transuranic exchange column dissolved in tri-n-butyl phosphate (TBP) and placed on a solid support column with 10 mL of 0.1 M HNO₃, and collect the eluent containing the rare earth elements plus americium fractions in beakers. Evaporate these solutions to dryness.

8.2.5 Dissolve the rare earth plus americium residues in 0.5 mL of 1.56 M HNO₃ in 80 % methanol (7.11).

8.2.6 Prepare a 2 mL column containing 200–400 mesh anion exchange resin.

8.2.7 Rinse with 0.3 M HNO₃ until the effluent solution tests negative for chloride ion using $AgNO_3$ solution (as evidenced by a lack of AgCl precipitate).

8.2.8 Condition the column with ~10 bed volumes of 1.56 M HNO₃ in 80 % methanol (7.11).

8.2.9 Transfer the solution containing the rare earth elements and americium to the column and discard the effluent to waste.

8.2.10 Once the loading solution reaches the top of the resin bed, rinse each beaker with 0.5 mL of loading solution and then transfer it to the appropriate column.

⁶ The sole source of supply of the apparatus known to the committee at this time is Eichrom Technologies LLC. Lisle, IL TRU Resin. If you are aware of alternative suppliers, please provide this information to ASTM International Headquarters. Your comments will receive careful consideration at a meeting of the responsible technical committee,¹ which you may attend.

🖽 C1769 – 15

8.2.11 Once the loading solution reaches the top of the resin bed, rinse each beaker with an additional 2.0 mL of loading solution, and then transfer it to the appropriate column.

8.2.12 Once the loading solution reaches the top of the resin bed, add two separate 1.0 mL portions of 0.156 M HNO₃ in 80% methanol (7.12) to the reservoir walls of each column.

8.2.13 Continue rinsing each column with an additional 19 mL of 0.156 M HNO₃ in 80 % methanol solution (7.12) to elute the elements heavier than neodymium.

8.2.14 Discard the eluent solution, and place a clean beaker under each column to collect the neodymium fraction.

8.2.15 Rinse the column with 15.0 mL of 0.156 M HNO₃ in 80 % methanol, then evaporate these neodymium fractions to dryness.

8.2.16 Submit the three purified U, Pu, and Nd fractions from each fuel aliquot for analysis by mass spectrometry analysis.

Note 1-Elution volumes needed to collect Nd fraction must be established for each laboratory column in conditions (resin volume and height).

9. Calculation

9.1 Calculate the heavy element atom percent fission from fission of ²³⁵U as follows:

$$F_{5} = N_{8}^{0} [(R_{5/8}^{0} - R_{5/8}) - (R_{6/8}^{0} - R_{6/8}^{0})]$$
(1)

9.2 Case I-At high exposure, where more than 10 % of the 235 U is consumed, calculate F_5 from Eq 1 assuming no loss of ²³⁸U or ²³⁶U during irradiation.

9.2.1 Eq 1 is extremely sensitive and adversely affected by natural uranium contamination in the irradiated sample, especially fully enriched uranium after short exposures. Ensure low uranium contamination is indicated by blank runs. Since 1 ng or more of environmental uranium may be introduced inadvertently into the sample during chemical separation, even after taking greatest care, samples processed for mass spectrometry should be larger than 50 μ g, where possible to minimize the effect of such contamination. The principal merit in Eq 1 lies in its independence of the reactor variable, a_5 . Eq 1 provides more accurate results than Eq 2 or Eq 3, if a_5 is known. Eq 1 is not recommended for use with recycled uranium fuel if R% is large with respect to $R_{\%}$.

9.3 *Case II*—At low exposures, calculate F_5 from Eq 2 and assume no loss of ²³⁶U during irradiation.

$$F_{5} = N_{8}^{0} \left[\left(R_{5/8}^{0} - R_{5/8} \right) - \left(R_{6/8}^{0} - R_{6/8}^{0} \right) \right]$$
(2)

Eq 2 is the most versatile of the three equations for F_5 . It is the equation most applicable to low exposures. However, it is equally valid for high exposures. Because the change in the ratio $R_{\%}$ is always greater than the corresponding change in the ratio $R_{\frac{5}{8}}$, Eq 2 is a more sensitive measure of F_5 than either Eq 1 or Eq 3. Furthermore, Eq 2 is 139 times less sensitive to natural uranium contamination than Eq 1 or Eq 3 because it is independent of ²³⁸U. Hence, where small amounts of natural uranium contaminate a ²³⁵U-enriched sample, use Eq 2. Eq 2 does require knowledge of a_5 . This value should be obtained from the reactor designer, by experiment, or machine calculation. Frequently, a_5 may not be known better than ± 4 to 20 % for any particular position in a given reactor. If a_5 is not otherwise available, see Figs. 1 and 2 where it is estimated as a function of neutron temperature for well-moderated thermal reactors from Refs (9) and (10). Values of a_5 for intermediateneutron-spectrum reactors increase markedly and can be as high as 0.65 (11). Values of a_5 for fast-neutron-spectrum reactors decrease again and can be as low as 0.09 (11). Eq 2 is not recommended for use with recycled U fuel if $R_{\%}^{0}$ is large with respect to $R_{\%}$.

9.4 Case III-For recycled U, if the initial ²³⁶U content is large and varies from piece to piece or is not known, calculate F_5 from Eq 3 and assume no loss of ²³⁸U during irradiation.

$$F_{5} = N_{8}^{0} [(R_{5/8}^{0} - R_{5/8}) / (1 + a_{5})]$$
(3)

Eq 3 also requires knowledge of a_5 . At small exposures, the equation involves the small difference between two numbers and is best applied where more than 10 % of the $^{235}\mathrm{U}$ is consumed. A significant bias can be introduced by natural U contamination in the irradiated sample, especially in fully enriched uranium after short exposure.

9.5 Calculate the atom percent fission due to ²³⁹Pu and ²⁴¹Pu from Eq 4-8 and assume no loss of ³⁸U or ²⁴²Pu during irradiation.

$$\Phi = (1 / \sigma_5 t) \ln(R_{5/8}^{0} / R_{5/8})$$
(4)

$$R'_{1/8} = R_{1/8} (\exp \lambda_1 t') + [1 + (\lambda_1 / \sigma_1 \Phi)][(1 + a_1)/a_1](R_{2/8}) - R_{2/8}(t)$$
(5)

$$P = \left(R_{0/8} - R_{0/8}^{0}\right) + \left(R'_{1/8} - R'_{1/8}^{0}\right)$$
(6)

$$F_{9} = N_{8}^{0} P/a_{9} \tag{7}$$

$$F_1 = N_8^{0} / a_1 (R_{2/8} - R_{2/8}^{0})$$
(8)

9.6 Calculate the atom percent fission due to ²³⁸U fast fission from Eq 9 and Eq 10 (5).

$$z = (\varepsilon - 1)[v_5 / (v_8 - 1 - a_8)] - 2.3(\varepsilon - 1)$$
(9)

$$F_8 = c\{F_5 + [(v_9 / v_5)/F_9] + [(v_1 / v_5)F_1]\}$$
(10)

9.7 Where accuracy in F_T of better than 10 % is required, correct all ratios after irradiation for burnout of ²³⁸U and ²³⁶U. Now that preliminary values of atom percent fission from various sources have been obtained, it becomes possible to estimate and correct for burnout of the 238 U and 236 U.

9.7.1 To correct for 238 U burnout, multiply $R_{5\%}$, $R_{5\%$ $R_{\frac{1}{8}}$, and $R_{\frac{2}{8}}$ by:

$$1/\{1 + R_{9/8} + P\left[(1 + a_9)/a_9\right] + (F_8 / N_8^0) - R_{9/8}^0\}$$
(11)

to obtain the corrected ratios. In this factor, the $R_{\%}$, P, and F_8 terms in the denominator are not corrected for ²³⁸U burnout on the first approximation. These corrections enter as the second order, the potential bias of neglecting them is small (for example, 0.01 %).

9.7.2 To correct for 236 U burnout, multiply R_{55} and the 238 U burnout-corrected $R_{5/8}$, by

$$\frac{\sigma_{5} - \sigma_{6}}{\sigma_{5}} \left\{ \frac{1 - R_{5/8} / R_{5/8}^{0}}{\exp[(\sigma_{6} \sigma_{5}) \ln(R_{5/8} / R_{5/8}^{0})] - R_{5/8} / R_{5/8}^{0}} \right\}$$
(12)

to obtain the corrected ratios. The $R_{5/8}$ used in this factor should be the 238 U-burnout-corrected ratio. The absorption cross sections for 235 U and 236 U, a_5 , and a_6 , are neutron spectrum dependent and must be obtained from the reactor designer, by experiment, or by machine calculation.

K

9.7.3 Recalculate F_5 , F_9 , F_1 , and F_8 from Eq 1-10 using the burnout-corrected values of all post-irradiation ratios.

9.8 Calculate total heavy element atom percent fission, F_T , from

$$F_{T} = F_{5} + F_{9} + F_{1} + F_{8} \tag{13}$$

9.9 Calculate the ratio of effective fission yields of 150 Nd to 148 Nd, $E_{^{50}\!/_{48}}$ as follows:

$$E_{50/48} = \left[R_{50/48} \left(R_{50/42} - C_{50/42} \right) \right] \left[R_{50/42} - R_{50/48} \left(C_{48/42} \right) \right]$$
(14)

where:

- $R_{50/48}$, $R_{50/42}$ = atom ratio of ¹⁵⁰Nd-to-¹⁴⁸Nd and ¹⁵⁰Nd-to-¹⁴²Nd in the unspiked sample, corrected for mass discrimination bias, and
- $C_{59/42}$, $C_{48/42}$ = atom ratios of ¹⁵⁰Nd-to-¹⁴²Nd and ¹⁴⁸Nd-to-¹⁴²Nd in natural neodymium contamination.

9.10 Calculate constants a, b, c, d, e and f as follows:

$$a = C_{42/50} - S_{42/50} \tag{15}$$

$$b = C_{48/50} - S_{48/50} \tag{16}$$

$$c = C_{42/50} S_{48/50} - S_{42/50} C_{48/50}$$
(17)

$$d = C_{42/50} \tag{18}$$

$$e = E_{50/48} C_{42/50} \tag{19}$$

$$f = \begin{pmatrix} 1 & - & E_{50/48} & C_{48/50} \end{pmatrix}$$
(20)

where:

- $C_{42/50}$, $C_{48/50}$ = atom ratio of ¹⁴²Nd and ¹⁴⁸Nd-to-¹⁵⁰Nd in natural neodymium contamination, which are 4.824 and 1.0195 respectively, and
- $S_{42/50}$, $S_{48/50}$ = atom ratio of ¹⁴²Nd and ¹⁴⁸Nd-to-¹⁵⁰Nd respectively in the spike solution.

9.11 Calculate $M'_{48/50}$ as follows:

$$M'_{48/50} = K(a \ M_{48/50} - b \ M_{42/50} - c)/(d - e \ M_{48/50} f \ M_{42/50})$$
(21)

where:

- factor to correct for a trace of non-fissioncaused ¹⁴²Nd from thermal neutron capture on ¹⁴⁷Nd found in Table 1. K is assumed to be unity for fast reactors,
- $M_{\frac{48}{50}}$ = atom ratio of fission product ¹⁴⁸Nd-to-spike ¹⁵⁰Nd adjusted for fission product ¹⁵⁰Nd, ¹⁴⁸Nd spike impurity, and ¹⁴⁸Nd and ¹⁵⁰Nd from natural neodymium contamination, and
- $M_{48/50}$, $M_{42/50}$ = measured atom ratio of ¹⁴⁸Nd-to-¹⁵⁰Nd and ¹⁴²Nd-to-¹⁵⁰Nd of the sample plus spike mixture corrected for mass discrimination bias (see 6.2).

9.12 Calculate the number of fissions per sample, F as follows:

$$F = (A_{50} / E_{48})M'_{48/50}$$
(22)

where:

 E_{48} = effective fractional fission yield of ¹⁴⁸Nd calculated from the fission yields of ¹⁴²Nd for each of the fissioning isotopes weighted according to their contribution to fission as measured in Test Method E244

The fractional yield for ¹⁴⁸Nd in thermal fission of ²³⁵U, ²³⁹Pu, and ²⁴¹Pu is 0.0167312 \pm 0.35 %, 0.016422 \pm 0.5 %, and 0.0193209 \pm 0.7 %, respectively; and for fast fission of ²³⁸U is 0.0209416 \pm 1.0 % (6), and

 A_{50} = the number of atoms of ¹⁵⁰Nd/mL of spike.

9.13 Calculate the atom fraction 238 U in the unspiked uranium sample, A_8 , as follows:

$$A_8 = R_{8:8} (R_{4:8} + R_{5:8} + R_{6:8} + R_{8:8})$$
(23)
where $R_{3:6}$ (which equals 1) is retained for clarity.

9.14 Calculate $S_{\frac{8}{3}}$ from $S_{\frac{3}{8}}$ as follows:

$$S_{8/3} = 1/S_{3/8} \tag{24}$$

9.15 Calculate the total uranium atoms per sample, U', from A_{33} :

$$U' = (A_{33} / A_8) \{ (M_{8/3} - S_{8/3}) / [1 - M_{8/3} / R_{8/3}] \}$$
(25)

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

Total Neutron Flux,	Total Neutron Exposure, fl (neutrons/cm ²)				
f (neutrons/cm ² /s)	1.3 10 ²⁰	3.3 10 ²⁰	1.3 10 ²¹	2.3 10 ²¹	3.3 10 ²¹
3 3 10 ¹²	0.9985	0.9985	0.9985	0.9985	0.9985
1 3 10 ¹³	0.9956	0.9952	0.9950	0.9985	0.9950
3 3 10 ¹³	0.9906	0.9870	0.9856	0.9853	0.9852
1 3 10 ¹⁴	0.9858	0.9716	0.9598	0.9569	0.9559
3 3 10 ¹⁴	0.9835	0.9592	0.9187	0.9008	0.8941
1 3 10 ¹⁵	0.9826	0.9526	0.8816	0.8284	0.8006

^AAssuming continuous reactor operation and a 274 6 91 barn ¹⁴⁷Nd effective neutron absorption cross section for a thermal neutron power reactor. This cross section was obtained by adjusting the 440 6 150 barn ¹⁴⁷Nd cross section (7) measured at 20°C to a Maxwellian spectrum at a neutron temperature of 300°C.

9.16 Calculate the atom fraction 239 Pu in the unspiked plutonium sample, A₉, as follows:

 $A_{9} = R_{9.9} / (R_{9.9} + R_{0.9} + R_{1.9} + R_{2.9})$ (26) where $R_{\%}$ (which equals 1) is retained for clarity.

9.17 Calculate $S_{\frac{5}{2}}$ from $S_{\frac{5}{2}}$ and $R_{\frac{5}{2}}$ from $R_{\frac{2}{3}}$ as follows:

$$S_{9/2} = 1/S_{2/9} \tag{27}$$

$$R_{9/2} = 1/R_{2/9} \tag{28}$$

9.18 Calculate the total plutonium atoms per sample, Pu', from A_{42} :

$$Pu' = A_{42} A_9 \{ (M_{9/2} - S_{9/2}) / [1 - (M_{9/2} / R_{9/2})] \}$$
(29)

9.19 Calculate the total heavy element atom percent fission, F_{τ} , from

$$F_{T} = \left[F' / \left(U' + P u' + F' \right) \right] \times 100$$
(30)

 $9.20\,$ If desired, calculate the gigawatt days per metric ton from

$$A = F_{\tau}(9.6 \pm 0.3) \tag{31}$$

9.21 Report heavy element atom percent fission or gigawatt days of heat per metric ton.

10. Precision and Bias

10.1 The precision and bias of atom percent fission obtained by this standard practice cannot be expressed by a single value.

Both are dependent upon many factors including the relative magnitude of the changes in isotopic abundances, the presence or absence of large amounts of preirradiation ²³⁶U, ²⁴⁰Pu, ²⁴¹Pu, and ²⁴²Pu, the certainty with which a_5 , a_9 , and a_1 are known for the particular neutron spectrum in which the fuel was irradiated, and the relative contributions of each fissionable nuclide to the total fissions. Each result must be individually evaluated.

10.2 Mass ratios can be obtained with a precision and bias of 1 %. The bias of F_5 , F_9 , F_1 , F_8 , and finally F_T can be obtained by propagation of errors of any sample. Although, F_5 varies from 2 to 10 relative percent depending upon the ²³⁵U burnup, F_9 is normally known for 10 to 20 % uncertainty in a_9 , which can differ for two positions of the same reactor. Lesser contributions from F_8 and F_1 are usually known to ±30 and ±60 %, respectively. The uncertainty of atom percent fission, F_T is approximately ±5 %, by summation of the sources of fission. A further error is introduced in converting U atom percent fission to gigawatt days per metric ton by the ±3 % uncertainty in the heat of fission.

11. Keywords

11.1 atom percent fission; burnup; fission product; mass spectrometric method; neutron flux; nuclear fuel; uranium and plutonium; uranium and plutonium fuel

REFERENCES

- Fudge, A. J., Wood, A. J., and Banham, M. F., "The Determination of Burnup in Nuclear Fuel Test Specimens Using Stable Fission Product Isotopes and Isotopic Dilution," USAEC Doc., TID-7629, 1961, pp. 152–165.
- (2) Rider, B. F., Peterson, Jr., J. P., and Ruiz, C. P., "Determination of Neodymium-148 in Irradiated UO₂ as a Measurement of Burnup," *Transactions of the American Nuclear Society*, Vol 7, No. 2, 1964, p. 350.
- (3) Garelis, E., "On the Roderick Fast Fission Calculation," USAEC Doc. GEAP-3595, 1960, p. 9.
- (4) Stehn, J. R., et al, "Neutron Cross Sections," Vol 3 "A=88 to 98," USAEC Doc. BNL-325, 2nd Ed., Supplement No. 2, February 1965.
- (5) Kouts, H., et al, "Exponential Experiments with Slightly Enriched Uranium Rods in Ordinary Water," Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, PIPAA, United Nations, Vol 5, 1956, p. 189.
- (6) Rider, B. F., and Ruiz, C. P., "Determination of Atom Per Cent Fission in Uranium Fuel," Progress in Nuclear Energy, Series IX, Analytical

Chemistry, Vol 2, Part 2, Crouthamel, C. E., ed, Pergamon Press, 1962.

- (7) Westcott, C. H., Walker, W. H., and Alexander, T. K., Proceedings of the International Conference in the Peaceful Uses of Atomic Energy, PIPAA, United Nations, Vol 16, 1958, p. 70.
- (8) Rider, B. F., et al "Determination of Uranium Burnup in MWD-Ton," USAEC Doc. GEAP-3373, 1960.
- (9) Rider, B. F., et al, "Accurate Nuclear Fuel Burnup Analysis XII," USAEC Doc., GEAP 4776, 1964.
- (10) Marsh, S. F., et al "Improved Two-Column Ion Exchange Separation of Plutonium, Uranium, and Neodymium in Mixed Uranium-Plutonium Fuels or Burnup Measurement," USAEC Doc. LA-5568, June 1975.
- (11) England, T. R., and Rider, B. F., "ENDF—349 Evaluation and Compliation of Fission Product Yields: 1993," Los Alamos National Laboratory, Los Alamos, NM, Report LA-UR-94–3106, October 1994.

ASTM International takes no position respecting the validity of any patent rights asserted in connection with any item mentioned in this standard. Users of this standard are expressly advised that determination of the validity of any such patent rights, and the risk of infringement of such rights, are entirely their own responsibility.

This standard is subject to revision at any time by the responsible technical committee and must be reviewed every five years and if not revised, either reapproved or withdrawn. Your comments are invited either for revision of this standard or for additional standards and should be addressed to ASTM International Headquarters. Your comments will receive careful consideration at a meeting of the responsible technical committee, which you may attend. If you feel that your comments have not received a fair hearing you should make your views known to the ASTM Committee on Standards, at the address shown below.

This standard is copyrighted by ASTM International, 100 Barr Harbor Drive, PO Box C700, West Conshohocken, PA 19428-2959, United States. Individual reprints (single or multiple copies) of this standard may be obtained by contacting ASTM at the above address or a 610-832-9585 (phone), 610-832-9555 (fax), or service@astm.org (e-mail); or through the ASTM website (www.astm.org). Permission rights to photocopy the standard may also be secured from the Copyright Clearance Center, 222 Rosewood Drive, Danvers, MA 01923, Tel: (978) 646-2600; http://www.copyright.com/