



# Standard Practice for Analysis of Spent Nuclear Fuel to Determine Selected Isotopes and Estimate Fuel Burnup<sup>1</sup>

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 ( $\epsilon$ ) 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 ( $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$ ,  $^{154}\text{Eu}$ ,  $^{106}\text{Ru}$ , and  $^{241}\text{Am}$ ). Fuel burnup is calculated from the  $^{148}\text{Nd}$ -to-fuel ratio as described in this method, which uses an effective  $^{148}\text{Nd}$  fission yield calculated from the fission yields of  $^{148}\text{Nd}$  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 reactor-dependent properties called for in the calculations (1, 2).<sup>2</sup>

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.*

<sup>1</sup> 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.

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<sup>2</sup> The boldface numbers in parentheses refer to a list of references at the end of this standard.

## 2. Referenced Documents

### 2.1 ASTM Standards:<sup>3</sup>

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)<sup>4</sup>

## 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  $F_5$ ,  $F_9$ ,  $F_I$ ,  $F_8$ —heavy element atom percent fission from fission  $^{235}\text{U}$ ,  $^{239}\text{Pu}$ ,  $^{241}\text{Pu}$ , and  $^{238}\text{U}$ .

3.3.2  $F_T$ —total heavy element atom percent fission.

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

3.3.4  $R_{5/8}^0$ ,  $R_{9/8}^0$ ,  $R_{5/5}^0$ —atoms ratios of  $^{235}\text{U}$  to  $^{238}\text{U}$ ,  $^{236}\text{U}$  to  $^{238}\text{U}$ , and  $^{236}\text{U}$  to  $^{235}\text{U}$  in the pre-irradiated fuel.

3.3.5  $R_{5/8}$ ,  $R_{9/8}$ ,  $R_{5/5}$ —atom ratios of  $^{235}\text{U}$  to  $^{238}\text{U}$ ,  $^{236}\text{U}$  to  $^{238}\text{U}$ , and  $^{236}\text{U}$  to  $^{235}\text{U}$  in the final irradiated sample.

3.3.6  $R_{5/8}$ ,  $R_{9/8}$ ,  $R_{5/5}$ —atom ratios of  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{241}\text{Pu}$ ,  $^{242}\text{Pu}$  and to  $^{238}\text{U}$  in the final irradiated sample.

<sup>3</sup> For referenced ASTM standards, visit the ASTM website, [www.astm.org](http://www.astm.org), or contact ASTM Customer Service at [service@astm.org](mailto:service@astm.org). For *Annual Book of ASTM Standards* volume information, refer to the standard's Document Summary page on the ASTM website.

<sup>4</sup> The last approved version of this historical standard is referenced on [www.astm.org](http://www.astm.org).

3.3.7  $R'_{1/8}$ —atom ratio of  $^{241}\text{Pu}$  to  $^{238}\text{U}$  in the final irradiated sample corrected for neutron capture, fission, and decay during and after irradiation.

3.3.8  $\nu_g$ — $2.67 \pm 0.30$  neutrons per fission of  $^{238}\text{U}$  (3).

3.3.9  $\nu_5$ — $2.426 \pm 0.006$  neutrons per fission of  $^{235}\text{U}$  (4).

3.3.10  $\nu_g/\nu_5$ —ratio of number of neutrons per fission of  $^{239}\text{Pu}$  to  $^{235}\text{U} = 1.192 \pm 0.005$  (4).

3.3.11  $\nu_f/\nu_5$ —ratio of number of neutrons per fission of  $^{241}\text{Pu}$  to  $^{235}\text{U} = 1.237 \pm 0.017$  (4).

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

3.3.13  $t$ —irradiation time, s.

3.3.14  $\lambda I$ —decay constant of  $153 \times 10^{-9} \text{ s}^{-1}$ .

3.3.15  $c$ —ratio of the  $^{238}\text{U}$  fission rate of the fission rate from all other sources expressed as equivalent  $^{235}\text{U}$  fission rate.

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

3.3.17  $a_5$ —effective ratio of  $^{235}\text{U}$  ( $n, \gamma$ ) 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  $a_g$ —effective ratio of  $^{239}\text{Pu}$  ( $n, \gamma$ ) 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  $a_f$ —effective ratio of  $^{241}\text{Pu}$  ( $n, \gamma$ ) 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  $a_g$ —effective ratio of  $^{238}\text{U}$  ( $n, \gamma$ ) capture-to-fission cross sections averaged over a fission spectrum =  $0.58 \pm 0.45$  (3).

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

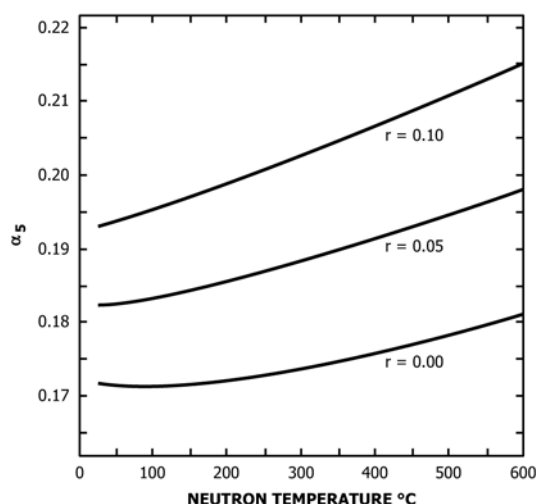


FIG. 1 Calculated Dependence of  $a_5$  on Neutron Temperature and Epithermal Index,  $r$ , for Well-Moderated Thermal Reactors

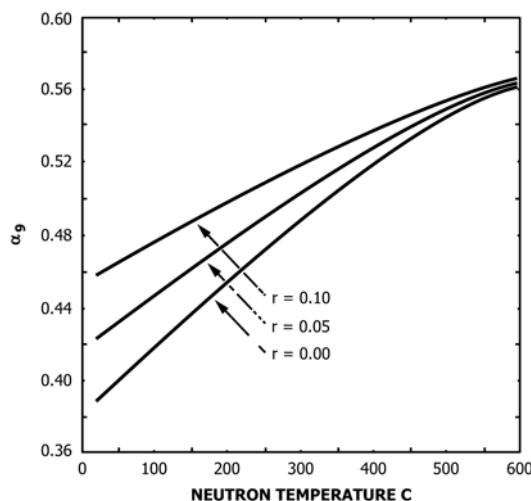


FIG. 2 Calculated Dependence of  $a_g$  on Neutron Temperature and Epithermal Index,  $r$ , for Well-Moderated Thermal Reactors

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

3.3.22  $\Phi$ —neutron flux, neutrons/cm<sup>2</sup>-s.

3.3.23  $\sigma_f, \sigma_s, \sigma_a$ —total neutron absorption cross sections of  $^{241}\text{Pu}$ ,  $^{235}\text{U}$ , and  $^{236}\text{U}$ . For boiling water reactors, typical core average values are  $188 \times 10^{-23} \text{ cm}^2$ ,  $64.6 \times 10^{-23} \text{ cm}^2$ , and  $5 \times 10^{-23} \text{ cm}^2$ , respectively. For pressurized water reactors, typical core average values are  $155 \times 10^{-23}$ ,  $55.6 \times 10^{-23} \text{ cm}^2$ , and  $8.4 \times 10^{-23} \text{ cm}^2$ , respectively.

3.3.24  $P$ —total  $^{239}\text{Pu}$  neutron captures per initial  $^{238}\text{U}$  atom.

#### 4. Summary of Practice

4.1 Atomic ratios of the isotopes  $^{234}\text{U}$ ,  $^{235}\text{U}$ ,  $^{236}\text{U}$ , to  $^{238}\text{U}$  and  $^{240}\text{Pu}$ ,  $^{241}\text{Pu}$ , and  $^{242}\text{Pu}$  to  $^{239}\text{Pu}$  are measured by mass spectrometry in accordance with Test Method C1625 or a similar methodology. The atom percent fission attributed to fission of  $^{235}\text{U}$ ,  $^{238}\text{U}$ ,  $^{239}\text{Pu}$ , and  $^{241}\text{Pu}$  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  $^{150}\text{Nd}$  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  $^{142}\text{Ce}$  or natural  $^{148}\text{Sm}$  and  $^{150}\text{Sm}$  is avoided by removing them in the chemical purification (9, 10).

4.3 After addition of a blended  $^{150}\text{Nd}$ ,  $^{233}\text{U}$ , and  $^{242}\text{Pu}$  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  $10^3$  and are normally limited to that value by traces of  $^{242}\text{Cm}$ ,  $^{147}\text{Pm}$ , and  $^{244}\text{Am}$ , respectively and

sometimes  $^{106}\text{Ru}$ ), and are insignificant to the analysis. The 70 ng  $^{148}\text{Nd}$  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  $^{148}\text{Nd}$  (for which a correction is made) without exceeding radiation dose rates of 20  $\mu\text{Sv/h}$  (20 mrem/h) at 1 m for 60-day cooled fuel to 20  $\mu\text{Sv/h}$  (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 ( $\frac{1}{2}$ -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  $\mu\text{g}$  run well in the mass spectrometer with both  $\text{NdO}^+$  and  $\text{Nd}^+$  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  $^{238}\text{U}$  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,  $\epsilon$ , 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  $^{235}\text{U}$  and  $^{236}\text{U}$  atom ratio content may vary from lot to lot.  $^{236}\text{U}$  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 examination of a large fuel element, the atom percent fission 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,  $^{148}\text{Nd}$  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 correc-

tions. (5) It has a low destruction cross section. (6) Formation of  $^{148}\text{Nd}$  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  $^{235}\text{U}$  and  $^{239}\text{Pu}$ . (9) Its fission yield is essentially independent of neutron energy (11). (10) It has a shielded isotope,  $^{142}\text{Nd}$ , which can be used for correcting natural neodymium contamination. (11) It is an atypical constituent of unirradiated fuel.

## 6. Apparatus

6.1 Dissolution bomb.<sup>5</sup>

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 ( $\text{HNO}_3$ ).

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 M  $\text{HNO}_3$ /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 M  $\text{HNO}_3$ /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  $\text{HNO}_3$ /80 %  $\text{CH}_3\text{OH}$ ) Add 6.3 mL concentrated nitric acid, 800 mL methanol and dilute, with water, to a final volume to 1000 mL volume with water.

<sup>5</sup> Parr dissolution bomb Model 4749 has shown to be adequate.