Designation: E 692 – 08

Standard Test Method for Determining the Content of Cesium-137 in Irradiated Nuclear Fuels by High-Resolution Gamma-Ray Spectral Analysis[1]

This standard is issued under the fixed designation E 692; 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 the number of atoms of ^{137}Cs in aqueous solutions of irradiated uranium and plutonium nuclear fuel. When combined with a method for determining the initial number of fissile atoms in the fuel, the results of this analysis allows atom percent fission (burn-up) to be calculated (**1**).[2] The determination of atom percent fission, uranium and plutonium concentrations, and isotopic abundances are covered in Test Methods E 267 and E 321.

1.2 ^{137}Cs is not suitable as a fission monitor for samples that may have lost cesium during reactor operation. For example, a large temperature gradient enhances ^{137}Cs migration from the fuel region to cooler regions such as the radial fuel-clad gap, or, to a lesser extent, towards the axial fuel end.

1.3 A nonuniform ^{137}Cs distribution should alert the analyst to the potential loss of the fission product nuclide. The ^{137}Cs distribution may be ascertained by an axial gamma-ray scan of the fuel element to be assayed. In a mixed-oxide fuel, comparison of the ^{137}Cs distribution with the distribution of non-migrating fission-product nuclides such as ^{95}Zr or ^{144}Ce would indicate the relative degree of ^{137}Cs migration.

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

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]

E 170 Terminology Relating to Radiation Measurements and Dosimetry
E 181 Test Methods for Detector Calibration and Analysis of Radionuclides
E 219 Test Method for Atom Percent Fission in Uranium Fuel (Radiochemical Method) (Discontinued 2001)[4]
E 267 Test Method for Uranium and Plutonium Concentrations and Isotopic Abundances
E 321 Test Method for Atom Percent Fission in Uranium and Plutonium Fuel (Neodymium-148 Method)

3. Summary of Test Method

3.1 ^{137}Cs is assayed by measuring the 662[5] keV gamma-ray emission rate from the isomeric transition of its metastable 2.6[6] min ^{137m}Ba daughter, using a high-resolution germanium detector and multichannel pulse-height analyzer. Refer to Test Methods E 181.

3.2 The number of atoms of ^{137}Cs in a sample is computed from the measured net gamma-ray count rate relative to the measured net gamma-ray count rate from a standard ^{137}Cs solution.

4. Significance and Use

4.1 This test method uses a high-resolution gamma-ray spectrometer as a basis for measuring the gamma-ray emission rate of ^{137}Cs-^{137m}Ba in a dilute nitric acid solution containing 10 mg/L of cesium carrier. No chemical separation of the cesium from the dissolved-fuel solution is required. The principal steps consist of diluting a weighed aliquot of the dissolved-fuel solution with a known mass of 1 *M* nitric acid (HNO_3) and measuring the 662 keV gamma-ray count rate from the sample, then measuring the 662 keV gamma-ray count rate from a standard source that has the same physical form and counting geometry as the sample.

4.2 The amount of fuel sample required for the analysis is small. For a sample containing 0.1 g of fuel irradiated to one

---

[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.

Current edition approved July 1, 2008. Published September 2008. Originally approved in 1979. Last previous edition approved in 2000 as E 692 – 00.

[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] Withdrawn. The last approved version of this historical standard is referenced on www.astm.org.

[5] The energy of the gamma ray is more precisely given in Ref (**2**) as 661.637 keV. For simplicity, all citations of this energy in this standard will be given as 662 keV.

[6] The half-life of this state is more precisely given in Ref (**3**) as 2.552 min. For simplicity, all citations of this half-life listed in this standard will be given as 2.6 min.

atom percent fission, a net count rate of approximately 10^5 counts per second will be observed for a counting geometry that yields a full-energy peak efficiency fraction of 1×10^{-3}. The advantage of this small amount of sample is that the concentration of fuel material can be kept at levels well below 1 g/L, which results in negligible self-absorption in the sample aliquot and a small radiation hazard to the analyst.

5. Precautions

5.1 Interferences from other gamma-ray emitting fission products are lessened by the use of a germanium detector with a minimum resolution of 3 keV full-width at half-maximum (FWHM) at 1332 keV, and by allowing four months or more for the sample to decay prior to measurement (**4**). Under these conditions, the gamma rays nearest to the 662 keV gamma ray of ^{137m}Ba will be the 637 keV gamma ray of ^{125}Sb and the 697 keV gamma ray of ^{144}Pr.

5.2 A slight complication of this test method is that the 662 keV gamma ray is superimposed on the Compton edge (defined in Terminology E 170) from the 766 keV gamma ray of ^{95}Nb and from the 796 keV gamma ray of ^{134}Cs, as shown in Fig. 1.

5.3 This test method requires accurately weighing an aliquot of the sample of fuel material containing sufficient ^{137}Cs-^{137m}Ba activity into a sample vial. In order to achieve the precision of which this test method is capable, the analyst should exercise great care when preparing the sample. To reduce the uncertainty associated with the sample quantity, aliquots should be prepared by weighing to an accuracy of better than 0.2 %. Weighing also reduces the calculation task, because in the case of burn-up analysis, the quantity of heavy-element atoms in the sample will have been determined on a mass-aliquot basis. The aliquot of sample solution should contain a weight of fuel sample of not less than 0.1 g weighed to an accuracy of 0.01 mg, and is to be diluted to the same total mass as the working standard. Uncertainties caused by slight variations in the counting-geometry among samples are negligible provided that the masses of the individual diluted samples are within ±0.01 g of each other.

5.4 The preparation of the ^{137}Cs reference standard should receive particular attention. Preferably, the number of ^{137}Cs atoms per gram of standard should have been determined by isotope-dilution mass spectrometry. An aliquot of not less than 0.1 g of the standard solution, weighed to ±0.1 mg, is diluted to total mass of 10.00 g with 1 *M* HNO_3 in a sample vial, which is then flame-sealed. A series of working standards with different concentrations should be prepared so that a working standard may be selected that will have approximately the same number of ^{137}Cs atoms as the sample to be measured.

6. Apparatus

6.1 *Germanium Detector*, with minimum resolution capability of 3 keV FWHM at 1332 keV, and associated electronics.

6.2 *Multichannel Pulse-Height Analyzer*, capable of a conversion ratio of 1 keV per channel (channel number versus gamma-ray energy in kiloelectronvolts).

7. Reagents and Materials

7.1 *Cesium-137 Standard Solution*—NIST SRM 4233E.[7]

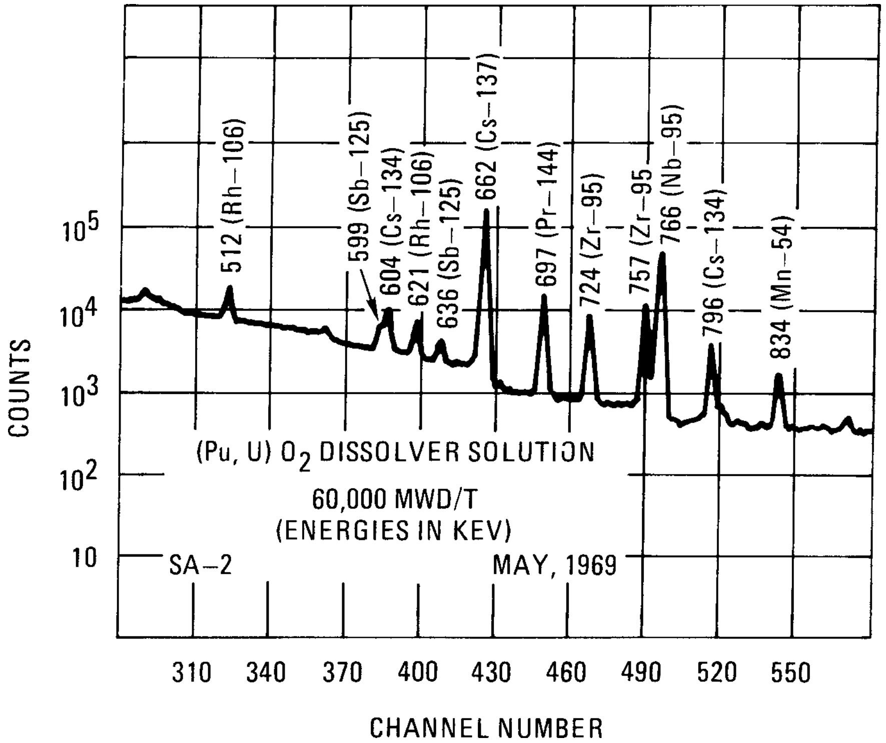

FIG. 1 Gamma-Ray Spectrum of a (Pu,U)O_2 Fuel, Irradiated to 6 Atom % Fission, and Decayed for Seven Months

---

[7] Available from the U.S. Department of Commerce, National Institute of Standards and Technology, Washington, DC 20234.

7.2 *Dilution Reagent*—1 *M* nitric acid (HNO_3), containing 10 mg/L of natural cesium carrier. Dissolve 15 mg of oven-dried (140°C) reagent-grade cesium nitrate ($CsNO_3$) per litre of 1 *M* HNO_3. Mix well.

7.3 *Vials*, flame-sealable, 20-mL standards ampoules.[8]

7.4 *Vials for Samples*, 20-mL nominal capacity.[9]

NOTE 1—Vials used for samples and standards should have the same dimensions.

8. Procedure

8.1 Weigh into a tared sample vial a sufficient aliquot of the solution containing the dissolved fuel to obtain 10 to 250 counts per second in the 662 keV peak. Weigh the sample mass to the nearest 0.1 mg and record.

8.2 Add sufficient 1 *M* HNO_3 to the sample vial to give a total (sample-plus-acid solution) mass of 10.00 ± 0.01 g.

8.3 Seal the vial with the cap and mix the solution by gentle swirling.

8.4 Place the sample vial in an exactly–reproducible position relative to the germanium detector so that a count rate of 10 to 250 s^{-1} in the 662 keV peak is obtained without exceeding a gross count rate of 3000 s^{-1}.

8.5 Accumulate sufficient counts in the 662 keV peak to achieve the desired precision. The standard deviation of the sample net peak counts is equal to the square root of the sum of the squares of the standard deviations of the counts associated with the Compton edge and background and with the gross peak counts. Thus, when the Compton and background counts comprise, for example, up to 50 % of the gross peak counts, then a total accumulation of 100 000 counts will be necessary to obtain a 1 % standard deviation. The net counts in the 662 keV peak should be greater than the Compton-background correction; otherwise one should use Test Method E 321, which involves the chemical separation of cesium from other radioactive interferences. The analyst must determine the optimum counting time needed to achieve the desired accuracy and precision.

8.6 Replace the sample vial with the standard, and count it to the same precision.

8.7 Determine the net counts in the 662 keV peak in both the sample spectrum and in the standard spectrum.

8.8 Use information provided on the ^{137}Cs Standard Solution, SRM 4233E, in conjunction with guidance from the supplier, NIST, to determine the ^{137}Cs atoms per gram contents in the standard sample.

8.9 Calculate the number of ^{137}Cs atoms in the sample from the ratio of the 662-ke V-peak countrates and the known number of ^{137}Cs atoms in the standard.

8.10 For determination of atom percent fission from the number of ^{137}Cs atoms in the sample, refer to Test Method E 219 and the current compilation of fission yields (**1**).

9. Precision and Bias

9.1 The estimated precision in an interlaboratory comparison with seven participating laboratories was 0.45 % relative standard deviation.

9.2 Two irradiated fuel sample solutions and a pure ^{137}Cs reference standard, each in a sealed ampoule, were circulated among the participating laboratories. The relative standard deviation for Sample 1, for a single laboratory was 0.40 % and for Sample 2 was 0.45 %. Sample 1 was from a uranium dioxide (UO_2) fuel irradiated to 3 atom % fission and decayed for 1.5 years. Sample 2 was from a mixed oxide $(Pu,U)O_2$ fuel irradiated to a 6 atom % fission and decayed for seven months. A portion of the gamma-ray spectrum of Sample 2 is shown in Fig. 1.

NOTE 2—Measurement uncertainty is described by a precision and bias statement in this standard. Another acceptable approach is to use Type A and Type B uncertainty components (**5**, **6**). The Type A/B uncertainty specification is now used in the International Organization for Standardization (ISO) standards and this approach can be expected to play a more prominent role in future uncertainty analyses.

10. Keywords

10.1 Cesium-137; gamma-ray spectrometer; nuclear fuel; percent fission

---

[8] The sole source of supply of the apparatus, vials, available as Item No. 176762, known to the committee at this time is Wheaton Glass Co., 1101 Wheaton Ave., Millville, NJ 08332. 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.

[9] The sole source of supply of the apparatus, Wheaton Crystal Lite glass liquid-scintillation counting vials, known to the committee at this time is Wheaton Glass Co., 1101 Wheaton Ave., Millville, NJ 08332. 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.

REFERENCES

(**1**) England, T. R., and Rider, B. F., *ENDF-349 Evaluation and Compilation of Fission Product Yields,* Los Alamos National Laboratory, Los Alamos, NM, report LA-UR-94-3106, ENDF-349, October 1994.

(**2**) "Evaluated Nuclear Structure Data File (ENSDF)," maintained by the National Nuclear Data Center (NNDC), Brookhaven National Laboratory, on behalf of the International Network for Nuclear Structure Data Evaluation, status as of May 3, 2008.

(**3**) "Nuclear Wallet Cards," National Nuclear Data Center (NNDC), Brookhaven National Laboratory, prepared by J. K. Tuli, April 2005.

(**4**) Matlack, G. M., Buzzelli, G., and Larsen, R. P., "Cs-137 Determination for Burnup Measurements by Gamma-Ray Spectrometry," Libby/Cockcroft Exchange Meeting on Burnup, LA-4430, Los Alamos Scientific Lab., Los Alamos, NM, June 9–11, 1969, Paper No. 37, p. 181.

(**5**) Taylor, B. N., Kuyatt, C. E., "Guidelines for Evaluating and Expressing the Uncertainty of NIST Measurement Results," *NIST Technical Note 1297*, National Institute of Standards and Technology, Gaithersburg, MD, 1994.

(**6**) "Guide to the Expression of Uncertainty in Measurement," International Organization for Standardization, 1995, ISBN 92-67-10188-9.

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 at 610-832-9585 (phone), 610-832-9555 (fax), or service@astm.org (e-mail); or through the ASTM website (www.astm.org).

Copyright ASTM International
Provided by IHS under license with ASTM
No reproduction or networking permitted without license from IHS

Licensee=Committee on Institutional Cooperation/5967164001
Not for Resale, 02/19/2012 15:35:01 MST