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AMERICAN SOCIETY FOR TESTING AND MATERIALS
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Standard Test Method for Atom Percent Fission in Uranium Fuel (Radiochemical Method)¹

This standard is issued under the fixed designation E 219; 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.

INTRODUCTION

Uranium atom percent fission in a nuclear fuel is frequently determined by measurement of a fission product to uranium (U) ratio in the irradiated U fuel **(1-7)**.²

Among radioactive fission products, cesium-137 (¹³⁷Cs) was among the first used for estimation of fuel burnup. It has a reasonably well-known decay scheme, a low capture cross section, a long half-life, a very low yield from ¹³⁶Cs by neutron capture, and a high-fission yield which is fairly accurately known and not greatly affected by the type of fissionable material or the neutron spectrum. Disadvantages are found in the volatility of Cs and its precursors at fuel operating temperature (which gives rise to migration within the fuel), interference in counting from ¹³⁴Cs and ¹³⁶Cs, some uncertainty in physical constants, and required knowledge of irradiation history.

Although this test method is of historical interest, test methods based on mass spectrometry are preferred as a basis for fuel-warranty settlements because they are more accurate.

1. Scope

1.1 This test method covers the determination of U atom percent fission that has occurred in U fuel from analysis of the ¹³⁷Cs to U ratio after irradiation.

1.2 The test method is applicable to high-density, clad U fuel (metal, alloy, ceramic compound) in which no separation of Cs and U has occurred.

1.3 The test method is best applied to fuels that have aged several months since irradiation. In such material, the 13-day ¹³⁶Cs activity is reduced to a small amount through decay **(3)**.

1.4 The test method should be restricted to low-exposure samples in which the activity of ¹³⁴Cs is less than that of ¹³⁷Cs. Cesium-134 is produced by neutron capture on fission product ¹³³Cs and grows at a rate proportional to the square of the exposure. This capture process limits the test method to samples exposed to less than 0.6×10^1 nvt. This exposure corresponds to burnups of 12 gigawatt days per metric ton of uranium (GWD/MTU) in Yankee Core I, and 5 GWD/MTU in Dresden Core I. Samples with higher exposures may require the use of a lithium-drifted germanium detector to obtain adequate resolution between ¹³⁴Cs and ¹³⁷Cs. The use of such a detector will extend the range of this test method by a factor of about 2. Mass spectrometric isotope dilution analysis of

¹³⁷Cs with ¹³³Cs as the isotopic diluent would also overcome ¹³⁴Cs interference.

1.5 The test method is best applied to fuels where overheating has not caused center melting or grain growth, since high temperatures cause ¹³⁷Cs to distill from the fuel and deposit on cooler regions, such as the cladding. Therefore, cladding should be leached in the dissolver solution or dissolved with the uranium to maintain the true ratio of fission product ¹³⁷Cs to U. Alternatively, burnup in a maximum flux region of a fuel element can be obtained from analyzing a low-flux region of the element. The burnup measured in this position can be related to the burnup in the peak-flux position by a gamma scan of the element. A gamma scan usually represents the distribution of zirconium-95 and is a reflection of the fission distribution integrated over only the most recent months. This is not always a serious disadvantage, since such studies may be made after short irradiations.

1.6 *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:

E 177 Practice for Use of the Terms Precision and Bias in ASTM Test Methods³

E 180 Practice for Determining the Precision of ASTM

¹ This test method is under the jurisdiction of ASTM Committee C-26 on Nuclear Fuel Cycle.

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² The boldface numbers in parentheses refer to the list of references appended to this test method.

³ Annual Book of ASTM Standards, Vol 14.02.

Methods for Analysis and Testing of Industrial Chemicals⁴

E 217 Test Method for Uranium by Controlled-Potential Coulometry⁵

E 267 Test Method for Uranium and Plutonium Concentrations and Isotopic Abundances⁵

E 318 Test Method for Uranium in Aqueous Solutions by Colorimetry⁶

E 320 Test Methods for Cesium-137 in Nuclear Fuel Solutions by Radiochemical Analysis⁵

E 321 Test Method for Atom Percent Fission in Uranium and Plutonium Fuel (Neodymium-148 Method)⁵

E 692 Test Method for Cesium-137 in Irradiated Nuclear Fuels by High-Resolution Gamma-Ray Spectral Analysis⁵

3. Terminology

3.1 Definitions:

3.1.1 *gigawatt days per metric ton*—the gigawatt days of heat produced per metric ton (megagram) of U initially present in a nuclear fuel.

3.1.2 *U atom percent fission*—the number of fissions per 100 U atoms initially present in a nuclear fuel.

4. Summary of Test Method

4.1 In principle, the number of fissions that have occurred in U fuel is determined by measuring the number of atoms of a fission product present and dividing by the fractional fission yield of that isotope. The fission yield of any fission product varies with the fissioning isotope, but a proper choice of fission product will be one that has a nearly constant yield for all fissionable isotopes and neutron spectra. The number of fissions occurring in ²³⁵U, ²³⁹Pu, and ²⁴¹Pu, as well as ²³⁸U, is thereby subject to measurement. The number of fissions, in turn, is divided by the atoms of U initially present. The resultant atom fraction, when multiplied by 100, yields U atom percent fission. This value can easily be converted, if desired, to gigawatt days per metric ton by multiplying by a constant.

5. Significance and Use

5.1 This test method provides a measure of U atom percent fission for U fuel, from which the output of heat during irradiation can be estimated.

5.2 The accuracy of this test method is limited, not only by the experimental error of the ¹³⁷Cs and U determinations, but also by the accuracy with which the fission yield and half-life of ¹³⁷Cs are known. The assumption is made that no separation of U from its fission products has occurred before withdrawing the sample for analysis. A simplifying assumption is made that the yield of ¹³⁷Cs from thermal fission of ²³⁵U is representative of all fissions occurring in U fuels. This assumption is only approximately true but introduces no greater uncertainty than many other uncertainties of the test method. A further simplifying assumption is made that the weight of initial ²³⁸U lost during the irradiation by formation of trans-U nuclides is negligible.

⁴ Annual Book of ASTM Standards, Vol 15.05.

⁵ Annual Book of ASTM Standards, Vol 12.02.

⁶ Annual Book of ASTM Standards, Vol 12.01.

6. Reagent

6.1 *Dilution Reagent*, consisting of nitric acid (1:1), containing 1 ppm of natural Cs carrier. Mix one volume of concentrated nitric acid (HNO₃, sp gr 1.42) with 1 volume of distilled water and dissolve therein 1.5 mg of reagent-grade cesium nitrate (CsNO₃) per liter.

NOTE 1—This amount of carrier will reduce the loss of ¹³⁷Cs by ion exchange with active centers in glassware. Losses of ¹³⁷Cs as high as 20 % during a 1-month period have been reported, whereas the activity of ¹³⁷Cs solutions containing 1 ppm of Cs carrier has been stable for several months (8).

7. Procedure

7.1 Prepare a working dilution of U dissolver solution (Note 2) with dilution reagent to obtain a final concentration of 100 to 1000 mg U/L.

NOTE 2—Dissolution procedures for fuels in common use are described elsewhere (9). With primary dissolver solutions high radiation levels are normally encountered. Adequate shielding should be provided and instrumentation for measurement of radiation should be on hand. Information as to the safe handling of radioactive materials is provided in National Institute of Standards and Technology *Handbooks* 42, 51, and 52, available from the U. S. Government Printing Office.

7.2 Obtain the ¹³⁷Cs concentration, *N*, in dilution in atoms per millilitre in accordance with Test Methods E 320 or Test Method E 692.

7.3 Obtain the U concentration in this dilution in milligrams of U per milliliter in accordance with Test Methods E 267, E 318, or E 217.

8. Calculation

8.1 Correct ¹³⁷Cs concentration for decay during and after irradiation as follows:

$$N' = N / \{e^{-\lambda t'}(1 - e^{-\lambda t})^{\lambda} t\} \quad (1)$$

where:

N' = atoms of ¹³⁷Cs per milliliter corrected for decay during and after irradiation,

N = ¹³⁷Cs atoms per milliliter at the time of measurement,

λ = ¹³⁷Cs decay constant = $7.32 \times 10^{-10} \text{ s}^{-1}$,

t' = elapsed time from the end of irradiation to measurement, s, and

t = irradiation time, s.

8.2 Calculate fissions per milliliter as follows:

$$F = N'/Y \quad (2)$$

where:

F = fissions per milliliter of solution,

N' = ¹³⁷Cs concentration corrected for decay during and after irradiation, atoms/mL, and

Y = ¹³⁷Cs fractional yield in thermal fission of ²³⁵U = 0.0623 (10).

8.3 Calculate the number of U atoms per milliliter as follows:

$$U = (C \times K)/M \quad (3)$$

where:

U = number of U atoms per milliliter of solution,

C = U concentration, mg/mL,