Standard Test Method for
Measuring Reaction Rates by Radioactivation of Uranium-238[1]

1. Scope

1.1 This test method covers procedures for measuring reaction rates by assaying a fission product (F.P.) from the fission reaction $^{238}U(n,f)F.P.$

1.2 The reaction is useful for measuring neutrons with energies from approximately 1.5 to 7 MeV and for irradiation times up to 30 to 40 years.

1.3 Equivalent fission neutron fluence rates as defined in Practice E261 can be determined.

1.4 Detailed procedures for other fast-neutron detectors are referenced in Practice E261.

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

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:*[2]

E170 Terminology Relating to Radiation Measurements and Dosimetry

E181 Test Methods for Detector Calibration and Analysis of Radionuclides

E261 Practice for Determining Neutron Fluence, Fluence Rate, and Spectra by Radioactivation Techniques

E262 Test Method for Determining Thermal Neutron Reaction Rates and Thermal Neutron Fluence Rates by Radioactivation Techniques

E320 Test Method for Cesium-137 in Nuclear Fuel Solutions by Radiochemical Analysis (Withdrawn 1993)[3]

E393 Test Method for Measuring Reaction Rates by Analysis of Barium-140 From Fission Dosimeters

E705 Test Method for Measuring Reaction Rates by Radioactivation of Neptunium-237

E844 Guide for Sensor Set Design and Irradiation for Reactor Surveillance, E 706 (IIC)

E944 Guide for Application of Neutron Spectrum Adjustment Methods in Reactor Surveillance, E 706 (IIA)

E1005 Test Method for Application and Analysis of Radiometric Monitors for Reactor Vessel Surveillance, E 706 (IIIA)

E1018 Guide for Application of ASTM Evaluated Cross Section Data File, Matrix E706 (IIB)

3. Terminology

3.1 *Definitions:*

3.1.1 Refer to Terminology E170.

4. Summary of Test Method

4.1 High-purity ^{238}U (<40 ppm ^{235}U) is irradiated in a fast-neutron field, thereby producing radioactive fission products from the reaction $^{238}U(n,f)F.P.$

4.2 Various fission products such as ^{137}Cs-^{137m}Ba, ^{140}Ba-^{140}La, ^{95}Zr, and ^{144}Ce can be assayed depending on the length of irradiation, purpose of the experiment, etc.

4.3 The gamma rays emitted through radioactive decay are counted, and the reaction rate, as defined in Practice E261, is calculated from the decay rate and the irradiation conditions.

4.4 The neutron fluence rate for neutrons with energies from approximately 1.5 to 7 MeV can then be calculated from the spectral-weighted neutron activation cross section as defined in Practice E261.

4.5 A parallel procedure that uses ^{237}Np instead of ^{238}U is given in Test Method E705.

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[1] This test method is under the jurisdiction of ASTM Committee E10 on Nuclear Technology and Applicationsand is the direct responsibility of Subcommittee E10.05 on Nuclear Radiation Metrology.

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[2] 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.

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

5. Significance and Use

5.1 Refer to Practice E261 for a general discussion of the determination of fast-neutron fluence rate with fission detectors.

5.2 ^{238}U is available as metal foil, wire, or oxide powder (see Guide E844). It is usually encapsulated in a suitable container to prevent loss of, and contamination by, the ^{238}U and its fission products.

5.3 One or more fission products can be assayed. Pertinent data for relevant fission products are given in Table 1 and Table 2.

5.3.1 ^{137}Cs-^{137m}Ba is chosen frequently for long irradiations. Radioactive products ^{134}Cs and ^{136}Cs may be present, which can interfere with the counting of the 0.662 MeV ^{137}Cs-^{137m}Ba gamma rays (see Test Methods E320).

5.3.2 ^{140}Ba-^{140}La is chosen frequently for short irradiations (see Test Method E393).

5.3.3 ^{95}Zr can be counted directly, following chemical separation, or with its daughter ^{95}Nb using a high-resolution gamma detector system.

5.3.4 ^{144}Ce is a high-yield fission product applicable to 2- to 3-year irradiations.

5.4 It is necessary to surround the ^{238}U monitor with a thermal neutron absorber to minimize fission product production from a quantity of ^{235}U in the ^{238}U target and from ^{239}Pu from (n,γ) reactions in the ^{238}U material. Assay of the ^{239}Pu concentration when a significant contribution is expected.

5.4.1 Fission product production in a light-water reactor by neutron activation product ^{239}Pu has been calculated to be insignificant (<2 %), compared to that from ^{238}U(n,f), for an irradiation period of 12 years at a fast-neutron (E > 1 MeV) fluence rate of 1×10^{11} cm^{-2} · s^{-1} provided the ^{238}U is shielded from thermal neutrons (see Fig. 2 of Guide E844).

TABLE 1 Recommended Nuclear Parameters for Certain Fission Products

Fission Product	Parent Half-Life^A (**1**)	Primary Radiation^A (**2**) (keV)	γ Probability of Decay^A (**2**)	Maximum Useful Irradiation Duration
^{95}Zr	64.032 (6) d	724.192 (4) 756.725 (12)	0.4427 (22) 0.5438	6 months
^{99}Mo	65.94 (1) h	739.500 (17) 777.921 (20)	0.1213 (22) 0.0426 (8)	300 hours
^{103}Ru	39.26 (2) d	497.085 (10)	0.910 (12)	4 months
^{137}Cs	30.05 (8) yr	661.657 (3)^B	0.8499 (20)^B	30–40 years
^{140}Ba – ^{140}La	12.7527 (23) d	537.261 (4) 1596.21 (4)	0.2439 (22) 0.9540 (8)^C 1.1515^D	1–1.5 months
^{144}Ce	284.91 (5) d	133.515 (2)	0.1109 (19)	2–3 years

^{A} The lightface numbers in parentheses are the magnitude of plus or minus uncertainties in the last digit(s) listed.
^{B} With ^{137m}Ba (2.552 min) in equilibrium.
^{C} Probability of daughter ^{140}La decay.
^{D} With ^{140}La (1.67855 d) in transient equilibrium.

TABLE 2 Recommended Fission Yields for Certain Fission Products^A

Fissile Isotope	Neutron Energy	Reaction Product	Type Yield	JEFF-3.1.1^{A,B} Fission Yield %
^{238}U(n,f)	0.5 MeV	^{95}Zr	RC	5.19 ± 1.714 %
		^{99}Mo	RC	6.18 ± 1.6 %
		^{103}Ru	RC	6.03 ± 1.6 %
		^{137}Cs	RC	6.02 ± 2.52 %
		^{137m}Ba	RI	1.0169e-2 ± 36.5 %
		^{140}Ba	RC	5.68 ± 2.67 %
		^{140}La	RI	6.8165e-6 ± 64 %
		^{144}Ce	RC	4.67 ± 2.46 %

^{A} The JEFF-3.1/3.1.1 radioactive decay data and fission yields sub-libraries, JEFF Report 20, OECD 2009, Nuclear Energy Agency.
^{B} All yield data given as a %; RC represents a cumulative yield; RI represents an independent yield.

5.4.2 Fission product production from photonuclear reactions, that is, (γ,f) reactions, while negligible near-power and research-reactor cores, can be large for deep-water penetrations (**3**).[4]

5.5 Good agreement between neutron fluence measured by ^{238}U fission and the ^{54}Fe(n,p)^{54}Mn reaction has been demonstrated (**4**). The reaction ^{238}U(n,f) F.P. is useful since it is responsive to a broader range of neutron energies than most threshold detectors.

5.6 The ^{238}U fission neutron spectrum-averaged cross section in several benchmark neutron fields is given in Table 3 of Practice E261. Sources for the latest recommended cross sections are given in Guide E1018. In the case of the ^{238}U(n, f)F.P. reaction, the recommended cross section source is the ENDF/B-VI release 8 cross section (MAT = 9237) (**5**). Fig. 1 shows a plot of the recommended cross section versus neutron energy for the fast-neutron reaction ^{238}U(n,f)F.P.

NOTE 1—The data is taken from the Evaluated Nuclear Data File, ENDF/B-VI, rather than the later ENDF/B-VII. This is in accordance with Guide E1018, Section 6.1, since the later ENDF/B-VII data files do not include covariance information. Some covariance information exists for ^{238}U in the standard sublibrary, but this is only for energies greater than 1 MeV. For more details, see Section H of Ref **6**.

6. Apparatus

6.1 *Gamma-Ray Detection Equipment* that can be used to accurately measure the decay rate of fission product activity are the following two types (**7**):

6.1.1 *NaI(T1) Gamma-Ray Scintillation Spectrometer* (see Test Methods E181 and E1005).

6.1.2 *Germanium Gamma-Ray Spectrometer* (see Test Methods E181 and E1005)—Because of its high resolution, the germanium detector is useful when contaminant activities are present.

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

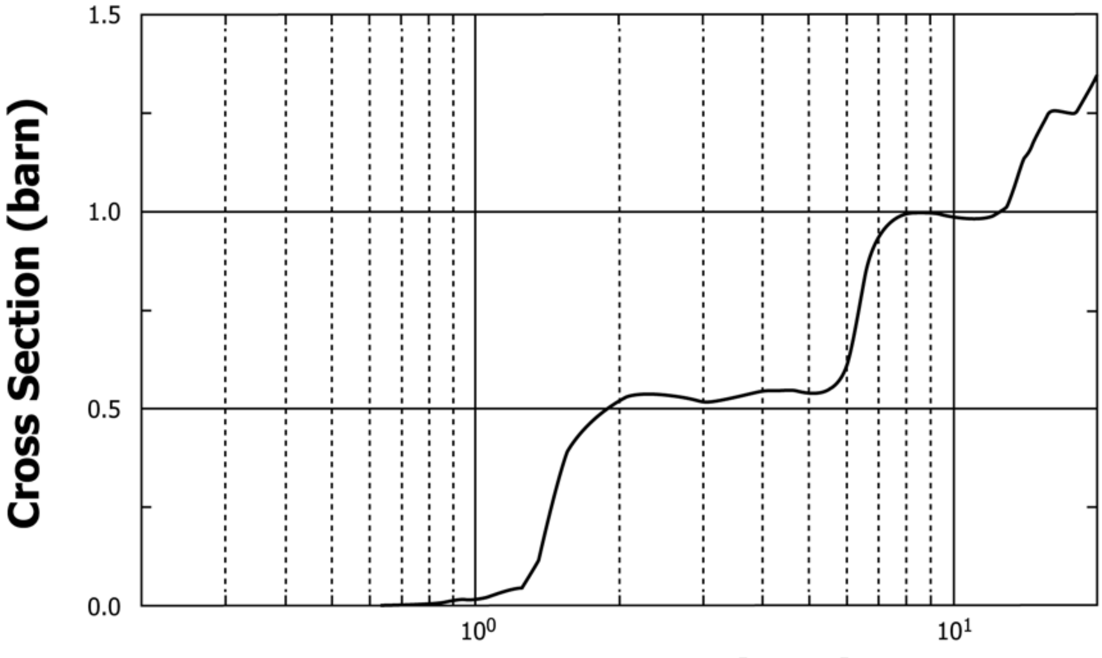

FIG. 1 ENDF/B-VI Cross Section Versus Energy for the $^{238}U(n,f)$F.P. Reaction

6.2 *Balance,* providing the accuracy and precision required by the experiment.

6.3 *Digital Computer,* useful for data analysis (optional).

7. Materials

7.1 *Uranium-238 Alloy or Oxide*—High-purity ^{238}U in the form of alloy wire, foil, or oxide powder is available.

7.1.1 The ^{238}U target material should be furnished with a certificate of analysis indicating any impurity concentrations.

7.2 *Encapsulating Materials*—Brass, stainless steel, copper, aluminum, quartz, or vanadium have been used as primary encapsulating materials. The container should be constructed in such a manner that it will not create significant perturbation of the neutron spectrum and fluence rate and that it may be opened easily, especially if the capsule is to be opened remotely. Certain encapsulation materials, for example, quartz and vanadium, allow gamma-ray counting without opening the capsule since there are no interfering activities.

8. Procedure

8.1 Select the size and shape of the sample to be irradiated, taking into consideration the size and shape of the irradiation space. The mass and exposure time are parameters that can be varied to obtain a desired count rate for a given neutron fluence rate.

8.2 Weigh the sample to the accuracy and precision required of the experiment; encapsulate; and, if irradiated in a thermal neutron environment, surround with a suitable high-melting thermal neutron absorber.

NOTE 2—The melting point of elemental cadmium is 321°C. For additional precautions, see Test Method E262.

8.3 Irradiate the sample for the predetermined time period. Record the power level and any changes in power during the irradiation, the time at the beginning and end of each power level, and the relative position of the monitors in the irradiation facility.

8.4 Check the sample for activity from cross contamination by other monitors or material irradiated in the vicinity or from any foreign substance adhering to the sample. Clean and reweigh, if necessary. If the sample is encapsulated oxide powder and if it is necessary to open the capsule, a suitable containment will be required.

8.4.1 If chemical separation is necessary, dissolution can be achieved in 8 *N* HNO_3-0.05 *N* HF.

NOTE 3—If an ion-exchange separation is to be subsequently performed, follow the dissolution by fuming with sulfuric acid (H_2SO_4) to expel fluorides. Fuming with H_2SO_4, however, may expel volatile fission product ruthenium, and, unless performed with care, losses of other fission products by spattering can occur.

8.5 Analyze the sample for fission-product content in disintegrations per second (see Test Methods E181, E320, and E1005).

8.5.1 It is assumed that the available apparatus has been calibrated to measure F.P. activity and that the experimenter is well versed in the operation of the apparatus.

8.5.2 Disintegration of ^{137}Cs nuclei produces 0.661657-MeV gamma rays with a probability per decay of 0.8210. It is recommended that a ^{137}Cs activity standard is used.

8.5.3 If the analyst is well versed in germanium counting and carefully calibrates the system, it is feasible to count ^{137}Cs-^{137m}Ba, ^{140}Ba-^{140}La, ^{95}Zr, and ^{144}Ce directly without chemical separation.

9. Calculation

9.1 Calculate the saturation activity, A_s, as follows:

$$A_s = A/y\left[\left(1 - \exp^{-\lambda ti}\right)\left(\exp^{-\lambda t_w}\right)\right] \quad (1)$$

where:

λ = disintegration constant for F.P., s^{-1},
A = number of disintegrations measured during the counting period, s^{-1},
t_i = irradiation duration, s,
t_w = elapsed time between the end of irradiation and counting, s, and
y = fission yield.

NOTE 4—This equation applies where transient equilibrium has been established, λ is that of the parent species. This equation should not be applied to the Ba/La line but can be applied to the other fission products. See Test Method E393 for reading the ^{140}Ba/^{140}La line.

NOTE 5—The equation for A_s is valid if the reactor operated at essentially constant power and if corrections for other reactions (for example, impurities, burnout, etc.) are negligible. Refer to Practice E261 for more generalized treatments.

9.2 Calculate the reaction rate,[5] R_s, as follows:

$$R_s = A_s/N_o \quad (2)$$

where:

N_o = number of target atoms.

9.3 Refer to Practice E261 and Guide E944 for a discussion of the determination of fast-neutron fluence rate and fluence.

10. Report

10.1 Practice E261 describes how data should be reported.

11. Precision and Bias

NOTE 6—Measurement uncertainty is described by a precision and bias statement in this standard. Another acceptable approach is to use Type A and B uncertainty components (**8, 9**). This Type A/B uncertainty specification is now used in International Organization for Standardization (ISO) Standards and this approach can be expected to play a more prominent role in future uncertainty analyses.

11.1 General practice indicates that disintegration rates can be determined with a bias of ± 5 % (1S %) and with a precision of ±1 % (1S %) (**10**).

11.2 The ^{238}U cumulative fission product yields have an uncertainty between 0.7 % and 7.7 % (1S %) for the various fission products as indicated in Table 1.

12. Keywords

12.1 fission dosimeter; fission product; fission reaction rates; Uranium-238

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[5] Within the context of this test method, the terms "fission rate" and "reaction rate" can be used synonymously.

REFERENCES

(1) *Nuclear Wallet Cards*, compiled by J. K. Tuli, National Nuclear Data Center, October 2011.

(2) Nuclear Data retrieval program NUDAT, a computer file of evaluated nuclear structure and radioactive decay data, which is maintained by the National Nuclear Data Center (NNDC), Brookhaven National Laboratory (BNL), on behalf of the International Network for Nuclear Structure Data Evaluation, which functions under the auspices of the Nuclear Data Section of the International Atomic Energy Agency (IAEA).

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(10) Adams, J. M., "Results from the NIST Round Robin Test of Fissionable Dosimeters in a Reactor Leakage Spectrum," *Reactor Dosimetry: Radiation Metrology and Assessment*, ASTM STP 1389, American Society for Testing and Materials, West Conshohocken, PA, 2001