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Standard Test Method for Measuring Reaction Rates by Analysis of Barium-140 From Fission Dosimeters[1]

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1. Scope

1.1 This test method describes two procedures for the measurement of reaction rates by determining the amount of the fission product ^{140}Ba produced by the non-threshold reactions ^{235}U (n, f), ^{241}Am (n, f), and ^{239}Pu (n, f), and by the threshold reactions ^{238}U (n, f), ^{237}Np (n, f), and ^{232}Th (n, f).

1.2 These reactions produce many fission products, among which is ^{140}Ba, having a half-life of 12.752 days. ^{140}Ba emits gamma rays of several energies; however, these are not easily detected in the presence of other fission products. Competing activity from other fission products requires that a chemical separation be employed or that the ^{140}Ba activity be determined indirectly by counting its daughter product ^{140}La. This test method describes both procedure (*a*), the nondestructive determination of ^{140}Ba by the direct counting of ^{140}La several days after irradiation, and procedure (*b*), the chemical separation of ^{140}Ba and the subsequent counting of ^{140}Ba or its daughter ^{140}La.

1.3 With suitable techniques, fission neutron fluence rates can be measured in the range from 10^7 n (neutrons) · cm^{-2}· s^{-1} to approximately 10^{15} n · cm^{-2} · s^{-1}.

1.4 The measurement of time-integrated reaction rates with fission dosimeters by ^{140}Ba analysis is limited by the half-life of ^{140}Ba to irradiation times up to about six weeks.

1.5 The values stated in SI units are to be regarded as 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:*

C 697 Test Methods for Chemical, Mass Spectrometric, and Spectrochemical Analysis of Nuclear-Grade Plutonium Dioxide Powders and Pellets[2]

D 1193 Specification for Reagent Water[3]

E 170 Terminology Relating to Radiation Measurements and Dosimetry[4]

E 181 Test Methods for Detector Calibration and Analysis of Radionuclides[4]

E 261 Practice for Determining Neutron Fluence Rate, Fluence, and Spectra by Radioactivation Techniques[4]

E 704 Test Method for Measuring Reaction Rates by Radioactivation of Uranium-238[4]

E 705 Test Method for Measuring Reaction Rates By Radioactivation of Neptunium-237[4]

E 844 Guide for Sensor Set Design and Irradiation for Reactor Surveillance, E706 (IIC)[4]

E 944 Guide for Application of Neutron Spectrum Adjustment Methods in Reactor Surveillance, (IIA)[4]

E 1005 Test Method for Application and Analysis of Radiometric Monitors for Reactor Vessel Surveillance, E706 (IIA)[4]

E 1018 Guide for Application of ASTM Evaluated Cross Section Data File, Matrix E 706 (IIB)[4]

3. Terminology

3.1 *Definitions:*

3.1.1 Refer to Terminology E 170.

4. Summary of Test Method

4.1 For nondestructive analysis, the fission dosimeter is allowed to cool for five days or more. The 1.596-MeV gamma energy peak of ^{140}La, which is the daughter product of the ^{140}Ba, is then counted. This information, combined with the decay constants for the La and the Ba, and the fission yield of the ^{140}Ba gives the reaction fission rate. When the proper cross section is used with the reaction rate, the equivalent fission fluence rate can be determined.

4.2 For destructive analysis, the fission product ^{140}Ba is separated from the irradiated fission dosimeter. The activity of the ^{140}Ba is determined by counting the 0.537 MeV gamma energy peak. This information is then used as in 4.1 to give the reaction rate or the equivalent fission fluence rate.

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

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[2] *Annual Book of ASTM Standards*, Vol 12.01.

[3] *Annual Book of ASTM Standards*, Vol 11.01.

[4] *Annual Book of ASTM Standards*, Vol 12.02.

5. Significance and Use

5.1 Refer to Guide E 844 for the selection, irradiation, and quality control of neutron dosimeters.

5.2 Refer to Practice E 261 for a general discussion of the measurement of neutron fluence rate and fluence. The neutron spectrum must be known in order to measure neutron fluence rates with a single detector. Also it is noted that cross sections are continuously being reevaluated. The latest recommended cross sections and details on how they can be obtained are discussed in Guide E 1018.

5.3 The reaction rate of a detector nuclide of known cross section, when combined with information about the neutron spectrum, permits the determination of the magnitude of the fluence rate impinging on the detector. Furthermore, if results from other detectors are available, the neutron spectrum can be defined more accurately. The techniques for fluence rate and fluence determinations are explained in Practice E 261.

5.4 ^{140}Ba is a radioactive nuclide formed as a result of uranium fission. Although it is formed in fission of any heavy atom, the relative yield will differ. Recommended fission yields for^{140}Ba production are given in Table 1. The direct (independent) fission yield of the daughter product ^{140}La, which is counted, is given in Table 2. These independent fission yields are relatively low compared to the ^{140}Ba cumulative fission yield and will not significantly affect the accuracy of the nondestructive procedure and need not be considered.

5.5 The half-life of ^{140}Ba is 12.752 days. Its daughter ^{140}La has a half-life of 1.6781 days.[5] The comparatively long half-life of^{140}Ba allows the counting to be delayed several weeks after irradiation in a high-neutron field. However, to achieve maximum sensitivity the daughter product ^{140}La should be counted five to six days after the irradiation during nondestructive analysis or five to six days after chemical separation if the latter technique is used. An alternative method after chemical separation is to count the ^{140}Ba directly.

5.6 Because of its 12.752 day half-life and substantial fission yield, ^{140}Ba is useful for irradiation times up to about six weeks in moderate intensity fields. One irradiation criterion is that the number of fissions produced should be approximately 10^9 or greater for good counting statistics. Also, if the irradiation time is substantially longer than six weeks the neutron fluence rate determined will apply mainly to the neutron field existing during the latter part of the irradiation. The ^{140}Ba decay constant and yield are known more accurately than those of many fission products, so it is sometimes used as a standard or base reaction with which other measurements can be normalized.

6. Apparatus

6.1 For nondestructive analysis the chemical separation equipment, materials, and reagents are not required.

6.2 *A NaI(Tl) or Germanium Gamma-Ray Spectrometer*, see Test Methods E 181 and E 1005.

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

TABLE 1 Recommended Cumulative Fission Yields for ^{140}Ba Production

Fission Dosimeter	Thermal or Fast Neutron Field	Fission Yield,%[A,B]
^{235}U	T	5.84596 ± 1 %
	F	5.98741 ± 1 %
^{238}U	F	5.84596± 1 %
^{239}Pu	T	5.31538± 1 %
	F	5.37475 ± 2 %
^{237}Np	F	5.47246 ± 1.4 %
^{232}Th	F	7.87647 ± 2.8 %
^{241}Am	T	5.95468 ± 2.8 %
	F	4.99172 ± 6 %

[A] These ENDF/B-VI values are considered the best available data. The uncertainties are expressed as a percentage of the fission yield.

[B] 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.

6.4 *Centrifuge*, clinical type, accommodating 50-mL centrifuge tubes.

6.5 *Steam Bath*.

6.6 *Ice Bath*.

6.7 *Drying Oven*.

6.8 *Filter Cones*.

6.9 *Fiberglass Filter Circles* for filter cone.

6.10 *Centrifuge Tubes*, 50-mL capacity.

6.11 *Fine Sintered-Glass Crucibles*.

TABLE 2 Independent Fission Yields for ^{140}La Production

Fission Dosimeter	Thermal or Fast Neutron Field	Fission Yield, %[A,B]
^{235}U	T	5.25214×10^{-3} ± 64 %
	F	1.31401×10^{-3} ± 64 %
^{238}U	F	1.38004×10^{-5} ± 64 %
^{239}Pu	T	8.11109×10^{-3} ± 64 %
	F	1.17572×10^{-2} ± 64 %
^{237}Np	F	4.421×10^{-3} ± 64 %
^{232}Th	F	2.71003×10^{-5} ± 64 %
^{241}Am	T	2.5758×10^{-2} ± 64 %
	F	2.07034×10^{-2} ± 32 %

[A] These ENDF/B-VI values are considered the best available data. The uncertainties are expressed as a percentage of the fission yield.

[B] 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.

7. Reagents and Materials

7.1 *Purity of Fission Dosimeters*—High purity uranium plutonium, neptunium, and thorium in the form of alloy wire, foil, or oxide powder are available.

7.1.1 *Target material* shall be furnished with a certificate of analysis indicating any impurity concentrations.

7.1.2 *Fission dosimeters* shall be encapsulated in hermetically sealed containers to avoid loss of materials and for health-hazard requirements.[6]

7.1.3 In *thermal reactors threshold reaction dosimeters* (for example, ^{238}U, ^{237}Np, ^{232}Th) shall be shielded from thermal neutrons with elemental, or compounds of, cadmium, gadolinium, or boron to prevent fission production from trace quantities (>40 ppm) of ^{235}U, and ^{239}Pu and to suppress

[5] *Nuclear Wallet Cards*, compiled by J. K. Tuli, National Nuclear Data Center, July 1990.

[6] Vanadium-encapsulated monitors of high purity are available from Isotope Sales Division, Oak Ridge National Laboratory, Oak Ridge, TN 37830.

buildup of interfering fissionable nuclides, for example, ^{239}Pu in the ^{238}U dosimeter, ^{238}Np and ^{238}Pu in the ^{237}Np dosimeter, and ^{233}U in the ^{232}Th dosimeter (see Guide E 844).

7.2 *Purity of Reagents*—Reagent grade chemicals shall be used in all tests. Unless otherwise indicated, it is intended that all reagents shall conform to the specifications of the Committee on Analytical Reagents of the American Chemical Society, where such specifications are available.[7] 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.

7.3 *Purity of Water*—Unless otherwise indicated, references to water shall be understood to mean reagent water as defined by Type II of Specification D 1193.

7.4 *Acetic Acid (36 %)*—Dilute 360 mL of glacial acetic acid to 1 L with water.

7.5 *Acetic Acid (6 %)*—Dilute 60 mL of glacial acetic acid to 1 L with water.

7.6 *Ammonium Acetate Solution (231 g/L)*—Dissolve 231 g of ammonium acetate in water and dilute to 1 L.

7.7 *Ammonium Hydroxide (sp gr 0.90)*—Concentrated ammonium hydroxide (NH_4OH).

7.8 *Barium Carrier (10 mg Ba/mL)*—See Section 8.

7.9 *Ethyl Alcohol* (95 %).

7.10 *Hydrochloric Acid (sp gr 1.42)*—Concentrated hydrochloric acid (HCl).

7.11 *Iron Carrier (10 mg Fe^{+++}/mL)*—Dissolve 48.4 g of $FeCl_3 \cdot 6H_2O$ in 100 mL of water and dilute to 1 L with water.

7.12 *Nitric Acid, Fuming*.

7.13 *Nitric Acid (sp gr 1.42)*—Concentrated nitric acid (HNO_3).

7.14 *Sodium Carbonate Solution*—Prepare a saturated solution of sodium carbonate (Na_2CO_3).

7.15 *Sodium Chromate Solution* (243 g/L)—Dissolve 243 g of sodium chromate (Na_2CrO_4) in water and dilute to 1 L.

7.16 *Strontium Holdback Carrier (10 mg Sr/mL)*—Dissolve 24.2 g of $Sr(NO_3)_2$ in 1 L of water. Mix well, filter through a glass wool, and store in a polyethylene bottle.

7.17 *Hydrofluoric Acid (HF) (1 N)*.

8. Preparation and Standardization of Barium Carrier

8.1 *Preparation and Standardization of Barium Carrier*:

8.1.1 Dissolve 19.0 g of barium nitrate ($Ba(NO_3)_2$) in deionized water and dilute to 1 L. Filter through glass wool and store in a polyethylene bottle.

8.2 *Standardization of Barium Carrier*:

8.2.1 Pipet 5.0 mL of the carrier solution into a 250-mL beaker and dilute to approximately 100 mL. Add 5 mL of acetic acid (36 %) and 10 mL of ammonium acetate solution. Bring to boiling; add 5 mL of Na_2CrO_4 solution dropwise with stirring; boil for 1 min with stirring. Cool the mixture to room temperature and filter the precipitated barium chromate ($BaCrO_4$) through a fine preweighed sintered-glass crucible.

8.2.2 Wash the precipitate three times with 5-mL portions of deionized water and three times with 5-mL portions of ethyl alcohol. Dry at 110°C, cool, and weigh. Calculate the barium content as follows:

$$Ba^{++}, \text{ mg/mL} = (W/V) \times 0.5421 \quad (1)$$

where:

W = milligrams of $BaCrO_4$, and
V = millilitres of carrier used.

9. Procedure for Nondestructive Analysis

9.1 Decide on the size and shape of sample to be irradiated (see Guide E 844).

9.2 Weigh the sample to the accuracy and precision of the experiment.

9.3 Place the sample in a cadmium, gadolinium, or boron cover if desired (see Guide E 844). Seal into a capsule when required by safety considerations.

9.4 Irradiate the sample for a predetermined period of time. Record the beginning and end of the irradiation period. Take into account any reactor power variation during the exposure period.

9.5 Prior to counting, remove any covering material from the dosimeter if it possesses interfering radionuclides. If encapsulated in quartz, copper, aluminum, or vanadium, the encapsulating material need not be removed before counting.

9.6 After five days after the irradiation, count the ^{140}La directly on a gamma-ray spectrometer (1.596-MeV gamma), or by coincidence counting.[8] Waiting exactly five days before counting is not required, but the ^{140}La is at its maximum about 134 h after the irradiation.

10. Procedure for Radiochemical Analysis

10.1 Decide on the size and shape of sample to be irradiated (see Guide E 844).

10.2 Weigh the sample to the accuracy and precision of the experiment.

10.3 Place the sample in a cadmium, gadolinium, or boron cover if desired (see Guide E 844). Seal into a capsule when required by safety considerations.

10.4 Irradiate the sample for a predetermined period of time. Record the beginning and end of the irradiation period. Take into account any reactor power variation during the exposure period. Since the fission product to be extracted, ^{140}Ba, has a 12.752-day half-life, there can be a several-day waiting period before the chemical separation is started.

10.5 Prior to counting, remove any covering material from the dosimeter. Dissolve the fission dosimeter in either 8 *N* HNO_3 to 0.05 *N* HF for uranium (see Test Method E 704), 12 *N* HNO_3–0.05 *N* HF for plutonium (see Test Methods C 697), 6 *N* HCl − 1 *N* HF for neptunium (see Test Method E 705), or concentrated HNO_3–0.01 *N* HF for thorium. Heat if necessary for complete dissolution.

10.6 Add 2 mL of standardized barium carrier. Transfer with a minimum amount of water into a 50-mL centrifuge tube.

[7] "Reagent Chemicals, American Chemical Society Specifications," Am. Chemical Soc., Washington DC. For suggestions on the testing of reagents not listed by the American Chemical Society, see "Reagent Chemicals and Standards," by Joseph Rosin, D. Van Nostrand Co., Inc., New York, NY, and the "United States Pharmacopeia."

[8] Dierckx, R., Maracci, G., and Rustichelli, F., "Measurement of the La-140 Fission Product Yield for Fissions in U-238 in a Thermal Reactor Type Spectrum," Journal of Nuclear Energy, Vol 25, pp. 85–89, 1971.

Neutralize the solution with saturated Na_2CO_3 solution; then add 5 mL in excess to precipitate barium carbonate ($BaCO_3$) and heat in a water bath. Centrifuge and discard the supernate. Dissolve the precipitate in a minimum of concentrated HNO_3. The total volume should not exceed 5 mL.

10.7 Add about 35 mL of fuming HNO_3 or enough to give a concentration of 7 parts fuming HNO_3 to 1 part sample. This step separates the barium and strontium from other activities; therefore the separation time should be recorded for counting.

10.8 Cool in an ice bath for 5 to 10 min and centrifuge, discard the supernate, and dissolve the precipitate with not more than 5 mL of water.

10.9 Repeat 10.7 and 10.8.

10.10 Dilute to 10 mL with water; add 10 drops of iron carrier. Mix and add concentrated NH_4OH dropwise until ferric hydroxide ($Fe(OH)_3$) precipitates; then add 10 drops in excess.

10.11 Centrifuge, transfer the supernate to a clean tube, and discard the precipitate. To the supernate, add 5 mL of saturated Na_2CO_3 solution. Heat in a water bath.

10.12 Centrifuge, discard the supernate, and dissolve the white precipitate in a *few drops* of concentrated HCl and a little water. Dilute to 10 mL; add 2.5 mL acetic acid (36 %) and 10 mL of ammonium acetate solution. Heat to boiling in water bath and add 2 mL of Na_2CrO_4 solution dropwise with constant swirling. Boil for 1 min and cool.

10.13 Centrifuge and discard the supernate. Dissolve the precipitate in 3 to 4 drops of concentrated HCl and a little water. Add 1 mL of strontium holdback carrier, dilute to 25 mL with water, and transfer to a 150-mL beaker. Add 1 mL of acetic acid (6 %) and heat to boiling. Then add 2 mL of ammonium acetate solution ($NH_4C_2H_3O_2$) dropwise with constant swirling followed by 1 to 2 drops of Na_2CrO_4 solution. Wash the precipitate on the filter with water and with ethyl alcohol. Dry the filter at 110°C for 10 min, cool to room temperature, weigh, and mount for counting.

NOTE 1—It may be necessary to heat the solution to dissolve the $BaCrO_4$. If the solution still remains cloudy, centrifuge and discard the precipitate, then add strontium holdback carrier, dilute to 25 mL, and continue with the next step.

10.14 Mount the filter paper and sample on a suitable holder for counting. A suggested method is to affix the sample to the center of a 2 by 3-in. (50 by 76-mm) card with double-backed tape, and cover with a thin mylar film.

10.15 Either count the barium precipitate (0.537-MeV gamma) as soon as possible after precipitation before the ^{140}La grows in or set it aside for about 134 h to allow the ^{140}La to reach a maximum before counting (1.596-MeV gamma), or both.

11. Calculation

11.1 Analyze the samples for ^{140}Ba or ^{140}La content in disintegrations per second with an apparatus listed in 6.2 (see Test Methods E 181 and E 1005).

11.1.1 It is assumed that the available apparatus has been calibrated and that the operator is well versed in its operation.

11.1.2 There are 0.2439 gammas of 537.261 keV per disintegration of ^{140}Ba and 0.954 gammas of 1596.21 keV per disintegration of ^{140}La.[9]

11.1.3 There are 8 gamma rays in coincidence with the 1.596-MeV gamma ray of ^{140}La. If the sample is placed close to the detector (10 cm or less), a correction factor should be applied to the measured peak counts (see Test Methods E 181).

11.1.4 No coincidence correction is necessary for the 0.537-MeV gamma ray of ^{140}Ba.

11.1.5 If the radiochemical analysis procedure was utilized, divide the measured disintegration rate by the chemical yield (weight of sample precipitate divided by the weight of the precipitate determined by standardization).

11.2 For nondestructive analysis calculate the reaction rate, *R*, from the equation:

$$R = \frac{A_1}{N\lambda_2\left\{\left[\frac{(Y_1)(1-e^{-\lambda 2ti})}{\lambda_2}+\frac{Y_1}{\lambda_2-\lambda_1}(e^{-\lambda 2ti}-e^{-\lambda 1ti})\right]e^{-\lambda 2tw}+\left[\frac{Y_1(1-e^{-\lambda 1ti})}{\lambda_2-\lambda_1}(e^{-\lambda 1tw}-e-\lambda_2{}^{tw})\right]\right.} \quad (2)$$

where:

R = reaction rate (fission per second per atom) of fission dosimeter in the sample,
A_1 = number of ^{140}La disintegrations per second at time t_w,
N = number of atoms of fission dosimeter,
Y_1 = fission yield of ^{140}Ba,
λ_1 = decay constant for $^{140}Ba = 6.29 \times 10^{-7}$ s^{-1},
λ_2 = decay constant for $^{140}La = 4.78 \times 10^{-6}$ s^{-1},
t_i = irradiation time, s, and
t_w = elapsed time between time of counting and irradiation, s.

NOTE 2—The terms "fission rate" and "reaction rate" can be used synonymously.

NOTE 3—The equation for *R* 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 E 261 for generalized treatments.

This rather long equation may be simplified by assuming that this $\lambda_1 t_i$, $\lambda_2 t_i < 1$. With this assumption the equation is as follows:

$$R = [A_1(\lambda_2 - \lambda_1)]/[N\lambda_2\ Y_1(e^{-\lambda 1tw} - e^{-\lambda 2tw})\ \lambda_1 t_i] \quad (3)$$

For short irradiation times (less than 1 h) this equation is accurate within 3 %.

11.3 For radiochemical analysis when the ^{140}Ba is counted immediately after separation the equation for *R* is as follows:

$$R = A_2/[NY_1(1 - e^{-\lambda 1ti})(e^{-\lambda 1tw})] \quad (4)$$

where:

A_2 = number of ^{140}Ba disintegrations per second.

11.4 For radiochemical analysis when the ^{140}La is counted several days after separation, the equation for *R* is as follows:

$$R = [A_1(\lambda_2 - \lambda_1)]/[\lambda_2 Y_1 N(e^{-\lambda 1ts} - e^{-\lambda 21ts}) \times (e^{-\lambda 1tw} - e^{-\lambda(ti+tw)})] \quad (5)$$

[9] 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).

where:
t_s = waiting time after separation, and
t_w = elapsed time between irradiation and separation.

For short irradiations ($\lambda_1 t_i < 1$) this can be simplified as follows:

$$R = [A_1(\lambda_2 - \lambda_1)e^{\lambda 1 tw}]/[\lambda_1\lambda_2 t_i Y_1 N \times (e^{-\lambda 1 ts} - e - \lambda_2{}^{ts})] \quad (6)$$

11.5 Refer to Practice E 261 and Guide E 944 for a discussion of the determination of fluence rate and fluence.

12. Precision and Bias

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

12.2 The fission product yield for ^{140}Ba has an uncertainty between 1 % and 6 % (1S %) for the various dosimeters as indicated in Table 1.

13. Keywords

13.1 Barium-140; fission dosimeter; fission product; fission reaction rates