Designation: E 705 – 96 (Reapproved 2002)

Standard Test Method for Measuring Reaction Rates by Radioactivation of Neptunium-237[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 ^{237}Np(n,f)F.P.

1.2 The reaction is useful for measuring neutrons with energies from approximately 0.7 to 6 MeV and for irradiation times up to 30 to 40 years.

1.3 Equivalent fission neutron fluence rates as defined in Practice E 261 can be determined.

1.4 Detailed procedures for other fast-neutron detectors are referenced in Practice E 261.

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:*

E 170 Terminology Relating to Radiation Measurements and Dosimetry[2]

E 181 Test Methods for Detector Calibration and Analysis of Radionuclides[2]

E 261 Practice for Determining Neutron Fluence Rate, Fluence, and Spectra by Radioactivation Techniques[2]

E 262 Test Method for Determining Thermal Neutron Reaction and Fluence Rates by Radioactivation Techniques[2]

E 320 Test Methods for Cesium-137 in Nuclear Fuel Solutions by Radiochemical Analysis[2]

E 393 Test Method for Measuring Reaction Rates by Analysis of Barium-140 from Fission Dosimeters[2]

E 482 Guide for Application of Neutron Transport Methods for Reactor Vessel Surveillance, E706 (IID)[2]

E 704 Test Method for Measuring Reaction Rates by Radioactivation of Uranium-238[2]

E 844 Guide for Sensor Set Design and Irradiation for Reactor Surveillance, E706 (IIC)[2]

E 944 Guide for Application of Neutron Spectrum Adjustment Methods in Reactor Surveillance, (IIA)[2]

E 1005 Test Method for Application and Analysis of Radiometric Monitors for Reactor Vessel Surveillance, E706 (IIIA)[2]

E 1018 Guide for Application of ASTM Evaluated Cross Section Data File, Matrix E 706 (IIB)[2]

3. Terminology

3.1 *Definitions:*

3.1.1 Refer to Terminology E 170.

4. Summary of Test Method

4.1 High-purity ^{237}Np (<40 ppm fissionable impurity) is irradiated in a fast-neutron field, thereby producing radioactive fission products from the reaction ^{237}Np(n,f)F.P.

4.2 Various fission products such as ^{137}Cs-^{137m}Ba, ^{1140}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 E 261, is calculated from the decay rate and the irradiation conditions.

4.4 The neutron fluence rate for neutrons with energies from approximately 0.7 to 6 MeV can then be calculated from the spectral-weighted neutron activation cross section as defined in Practice E 261.

4.5 A parallel procedure that uses ^{238}U instead of ^{237}Np is given in Test Method E 704.

5. Significance and Use

5.1 Refer to Practice E 261 for a general discussion of the determination of fast-neutron fluence rate with fission detectors.

5.2 ^{237}Np is available as metal foil, wire, or oxide powder. For further information, see Guide E 844. It is usually encapsulated in a suitable container to prevent loss of, and contamination by, the ^{237}Np and its fission products.[3]

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

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[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 12.02.

[3] Vanadium-encapsulated monitors of high purity are available from the Isotope Sales Div., Oak Ridge, TN 37830.

TABLE 1 Recommended Nuclear Parameters for Certain Fission Products

Fission Product	Parent Half-Life^A **(6)**	Primary Radiation^A **(7)** (keV)	γ Probability of Decay^A **(7)**	Maximum Useful Irradiation Duration
^{95}Zr	64.04 (4) d	724.199 (5) 756.729 (12)	0.4417 (19) 0.5446	6 months
^{99}Mo	65.94 (1) h	0.1213 0.0435	739.5 777.921	300 hours
^{103}Ru	39.254 (8) d	497.084 (10)	0.910 (23)	4 months
^{137}Cs	30.0 (2) yr	661.660 (3)^B	0.8510^B	30–40 years
^{140}Ba–^{140}La	12.746 (10) d	537.31 (4) 1596.54 (14)	0.2439 0.9540^C 1.1515^D	1–1.5 months
^{144}Ce	284.9 (2) d	133.515 (8)	0.1109 (4)	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 (1.6780 d) in transient equilibrium.

TABLE 2 Recommended Fission Yields for Certain Fission Products^A

Fissile Isotope	Neutron Energy	Reaction Product	Type Yield	ENDF/B-VI^{B,A} Fission Yield (%)
^{237}Np(n,f)	0.5 MeV	^{95}Zr	RC	5.68896 ± 2.8 %
		^{99}Mo	RC	6.11547 ± 4 %
		^{103}Ru	RC	5.56212 ± 2.8 %
		^{137}Cs	RC	6.16977 ± 2.8 %
		^{137m}Ba	RI	1.438e-5 ± 64 %
		^{140}Ba	RC	6.17171 ± 2 %
		^{140}La	RI	4.421e-5 ± 64 %
		^{144}Ce	RC	4.12987 ± 2 %

^{A}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.

^{B}All yield data given as a %; RC represents a cumulative yield; RI represents an independent yield.

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 ray (see Test Methods E 320).

5.3.2 ^{140}Ba-^{140}La is chosen frequently for short irradiations (see Test Method E 393).

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 ^{237}Np monitor with a thermal neutron absorber to minimize fission product production from trace quantities of fissionable nuclides in the ^{237}Np target and from ^{238}Np and ^{238}Pu from (n,γ) reactions in the ^{237}Np material. Assay of ^{238}Pu and ^{239}Pu concentration is recommended when a significant contribution is expected.

5.4.1 Fission product production in a light-water reactor by neutron activation products ^{238}Np and ^{238}Pu has been calculated to be insignificant (1.2 %), compared to that from ^{237}Np(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 ^{237}Np is shielded from thermal neutrons (see Fig. 2 of Guide E 844).

5.4.2 Fission product production from photonuclear reactions, that is, (γ,f) reactions, while negligible near-power and researchreactor cores, can be large for deep-water penetrations **(1).**[4]

5.5 Good agreement between neutron fluence measured by ^{237}Np fission and the ^{54}Fe(n,p)^{54}Mn reaction has been demonstrated **(2).** The reaction ^{237}Np(n,f) F.P. is useful since it is responsive to a broader range of neutron energies than most threshold detectors.

5.6 The ^{237}Np fission neutron spectrum-averaged cross section in several benchmark neutron fields are given in Table 3 of Practice E 261. Sources for the latest recommended cross sections are given in Guide E 1018. In the case of the ^{237}Np(n,f)F.P. reaction, the recommended cross section source is the ENDF/B-VI cross section (MAT = 9346) revision 1 **(3)**. Fig. 1 shows a plot of the recommended cross section versus neutron energy for the fast-neutron reaction ^{237}Np(n,f)F.P.

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 **(4):**

6.1.1 *NaI(T1) Gamma-Ray Scintillation Spectrometer* (see Test Methods E 181 and E 1005).

6.1.2 *Germanium Gamma-Ray Spectrometer* (see Test Methods E 181 and E 1005)—Because of its high resolution, the germanium detector is useful when contaminant activities are present.

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

6.3 *Digital Computer*, useful for data analysis, but is not necessary (optional).

7. Materials

7.1 *Neptunium-237 Alloy or Oxide*—High-purity ^{237}Np in the form of alloy wire, foil, or oxide powder is available.

7.1.1 The ^{237}Np target material should be furnished with a certificate of analysis indicating any impurity concentrations.

7.2 *Encapsulating Materials*—Brass, stainless steel, copper, aluminum, vanadium, and quartz 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 or 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.

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

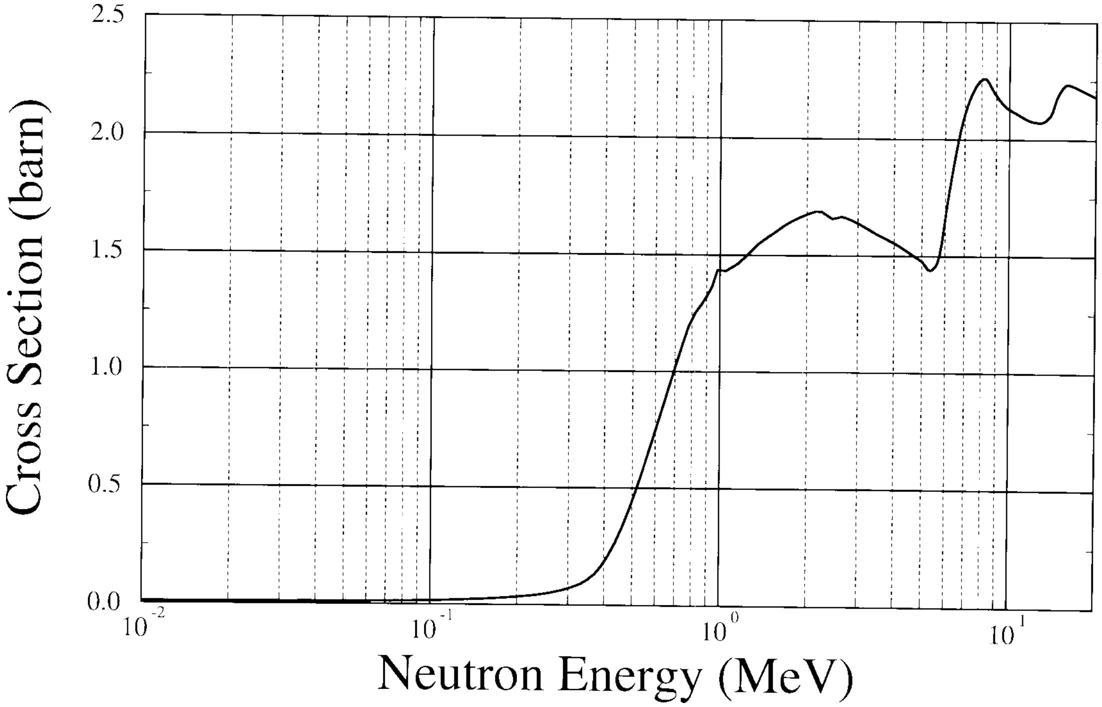

FIG. 1 ENDF/B-VI Cross Section Versus Energy for the $^{237}Np(n,f)F.P.$ Reaction

NOTE 1—The melting point of elemental cadmium is 321°C. For additional precautions, see Test Method E 262.

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, suitable containment will be required.

8.4.1 If chemical separation is necessary, dissolution can be achieved in 6 *N* HCl-1 *N* HF with periodic additions of H_2O_2, followed by fuming with H_2SO_4.

NOTE 2—Fuming with H_2SO_4 may expel volatile fission product ruthenium and, unless performed with care, losses of other fission products by spattering can occur.

NOTE 3—A specific dissolution procedure has been developed for vanadium-encapsulated neptunium oxide specimens **(5).**

8.5 Analyze the sample for fission-product content in disintegrations per second (see Test Methods E 181, E 320, and E 1005).

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 0.662-MeV gamma rays with a probability per decay of 0.852. 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:

$$A_s = A/y[(1 - exp - \lambda t_i)(exp - \lambda t_w)] \quad (1)$$

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—Where transient equilibrium has been established, λ is that of the parent species.

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 E 261 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 E 261 and Guide E 944 for a discussion of the determination of fast neutron fluence rate.

10. Report

10.1 Practice E 261 describes how data should be reported.

11. Precision and Bias

11.1 General practice indicates that disintegration rates can

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[5] The terms "fission rate" and "reaction rate" can be used synonymously.

be determined with a bias of ±5 % (1S %) and with a precision of ±1 % (1S %).

11.2 The ^{237}Np cumulative fission product yields have an uncertainty between 2 % and 4 % (1S %) for the various fission products as indicated in Table 1.

12. Keywords

12.1 fission dosimeter; fission product; fission reaction rates; Neptunium-237

REFERENCES

(**1**) Verbinski, V. V., et al, "Measurements and Calculations of Photofission Effects in a Swimming Pool Type Reactor," *Transactions of the American Nuclear Society*, Washington, DC, Vol 30, November 1978.

(**2**) Barry, K. M., and Corbett, J. A., "Measurement of Neutron Fluence by Neptunium-237 and Uranium-238 Fission Dosimeters," *Nuclear Technology*, Vol 11, May 1971.

(**3**) ENDF/B-V Dosimetry Tape 531-G, Mat. No. 6399 (93-Np-237), October 1979.

(**4**) Crouthamel, C. E. (Revised by Adams, F., and Dams, R.), *Applied Gamma-Ray Spectrometry*, Pergamon Press, 1970.

(**5**) "ENDF-201, ENDF/B-VI Summary Documentation," P. F. Rose, Ed. Brookhaven National Laboratory Report BNL-NCS-174, 4th Edition, October, 1991.

(**6**) *Nuclear Wallet Cards*, compiled by J. K. Tuli, National Nuclear Data Center, July 1990.

(**7**) 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).