



Designation: E705 – 18

# Standard Test Method for Measuring Reaction Rates by Radioactivation of Neptunium-237<sup>1</sup>

This standard is issued under the fixed designation E705; 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 ( $\epsilon$ ) indicates an editorial change since the last revision or reapproval.

## 1. Scope

1.1 This test method covers procedures for measuring reaction rates by assaying a fission product (F.P.) from the fission reaction  $^{237}\text{Np}(n,f)\text{F.P.}$

1.2 The reaction is useful for measuring neutrons with energies from approximately 0.7 to 6 MeV and for irradiation times up to 90 years, provided that the analysis methods described in Practice E261 are followed. If dosimeters are analyzed after irradiation periods longer than 90 years, the information inferred about the fluence during irradiation periods more than 90 years before the end of the irradiation should not be relied upon without supporting data from dosimeters withdrawn earlier.

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, health, and environmental practices and determine the applicability of regulatory limitations prior to use.*

1.7 *This international standard was developed in accordance with internationally recognized principles on standardization established in the Decision on Principles for the Development of International Standards, Guides and Recommendations issued by the World Trade Organization Technical Barriers to Trade (TBT) Committee.*

<sup>1</sup> 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. Referenced Documents

2.1 *ASTM Standards:*<sup>2</sup>

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)<sup>3</sup>

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

E704 Test Method for Measuring Reaction Rates by Radioactivation of Uranium-238

E844 Guide for Sensor Set Design and Irradiation for Reactor Surveillance

E944 Guide for Application of Neutron Spectrum Adjustment Methods in Reactor Surveillance

E1005 Test Method for Application and Analysis of Radiometric Monitors for Reactor Vessel Surveillance

E1018 Guide for Application of ASTM Evaluated Cross Section Data File

## 3. Terminology

3.1 *Definitions:*

3.1.1 Refer to Terminology E170.

## 4. Summary of Test Method

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

<sup>2</sup> 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.

<sup>3</sup> The last approved version of this historical standard is referenced on www.astm.org.

4.2 Various fission products such as  $^{137}\text{Cs}$ - $^{137\text{m}}\text{Ba}$ ,  $^{140}\text{Ba}$ - $^{140}\text{La}$ ,  $^{95}\text{Zr}$ , and  $^{144}\text{Ce}$  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 0.7 to 6 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  $^{238}\text{U}$  instead of  $^{237}\text{Np}$  is given in Test Method E704.

## 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  $^{237}\text{Np}$  is available as metal foil, wire, or oxide powder. For further information, see Guide E844. It is usually encapsulated in a suitable container to prevent loss of, and contamination by, the  $^{237}\text{Np}$  and its fission products.<sup>4</sup>

5.3 One or more fission products can be assayed. Pertinent data for relevant fission products are given in Table 1<sup>5</sup> and Table 2.

<sup>4</sup> The sole source of supply of Vanadium-encapsulated monitors of high purity known to the committee at this time in the United States is the National Isotope Development Center, Isotope Business Office, Oak Ridge National Laboratory, Oak Ridge, TN 37830. In Europe, the sole source of supply is European Commission, JRC, Institute for Reference Materials and Measurements (IRMM) Reference Materials Unit Retieseweg 111, B-2440 Geel, Belgium. 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,<sup>1</sup> which you may attend.

<sup>5</sup> The boldface numbers in parentheses refer to the list of references appended to this test method.

TABLE 2 Recommended Fission Yields for Certain Fission Products<sup>A</sup>

Fissile Isotope	Neutron Energy <sup>C</sup>	Reaction Product	Type Yield	JEFF 3.1.1 <sup>B,A</sup> Fission Yield (%)
$^{237}\text{Np}(n,f)$	0.4 MeV	$^{95}\text{Zr}$	RC	5.6147 ± 2.7 %
		$^{99}\text{Mo}$	RC	7.6218 ± 16.304 %
		$^{103}\text{Ru}$	RC	5.43050 ± 12.7 %
		$^{137}\text{Cs}$	RC	6.26540 ± 3.71 %
		$^{137\text{m}}\text{Ba}$	RI	1.48020e-3 ± 35.58 %
		$^{140}\text{Ba}$	RC	5.73800 ± 2.3 %
		$^{140}\text{La}$	RI	6.35680e-3 ± 36.68 %
		$^{144}\text{Ce}$	RC	4.12300 ± 4.7 %

<sup>A</sup> The JEFF-3.1/3.1.1 radioactive decay data and fission yields sub-libraries, JEFF Report 20, OECD 2009, Nuclear Energy Agency (2).

<sup>B</sup> All yield data given as a %; RC represents a cumulative yield; RI represents an independent yield.

<sup>C</sup> The neutron energy represents a generic "fast neutron" spectrum and has been characterized in the JEFF 3.1.1 fission yield library as having an average neutron energy of 0.4 MeV.

5.3.1  $^{137}\text{Cs}$ - $^{137\text{m}}\text{Ba}$  is chosen frequently for long irradiations. Radioactive products  $^{134}\text{Cs}$  and  $^{136}\text{Cs}$  may be present, which can interfere with the counting of the 0.661657 MeV  $^{137}\text{Cs}$ - $^{137\text{m}}\text{Ba}$  gamma ray (see Test Methods E320).

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

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

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

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

5.4.1 Fission product production in a light-water reactor by neutron activation products  $^{238}\text{Np}$  and  $^{238}\text{Pu}$  has been calculated to be insignificant (1.2 %), compared to that from  $^{237}\text{Np}(n,f)$ , for an irradiation period of 12 years at a fast neutron ( $E > 1$  MeV) fluence rate of  $1 \times 10^{11} \text{ cm}^{-2} \cdot \text{s}^{-1}$ , provided the  $^{237}\text{Np}$  is shielded from thermal neutrons (see Fig. 2 of Guide E844).

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).

5.5 This dosimetry reaction is important in the area of reactor retrospective dosimetry (4, 5). Good agreement between neutron fluence measured by  $^{237}\text{Np}$  fission and the  $^{54}\text{Fe}(n,p)^{54}\text{Mn}$  reaction has been demonstrated (6, 7). The reaction  $^{237}\text{Np}(n,f)$  F.P. is useful since it is responsive to a broader range of neutron energies than most threshold detectors.

5.5.1 Fig. 1 shows the energy-dependent cross section for this dosimetry reaction. The figure shows that, while it is not strictly a threshold detector, because of its sensitivity in the greater than 0.1 MeV neutron energy range it can function as

TABLE 1 Recommended Nuclear Parameters for Certain Fission Products

Fission Product	Parent Half-Life <sup>A,ε(1)</sup>	Primary Radiation <sup>A,ε(1)</sup> (keV)	γ Probability of Decay <sup>A,ε(1)</sup>	Maximum Useful Irradiation Duration
$^{95}\text{Zr}$	64.032 (6) days	724.193 (3) 756.729 (12)	0.4427 (22) 0.5438 (22)	6 months
$^{99}\text{Mo}$	2.747 (6) days	739.500 (17) 777.921 (20)	0.122 (15) 0.0428 (8)	300 h
$^{103}\text{Ru}$	39.247 (13) days	497.085 (10)	0.910 (12)	4 months
$^{137}\text{Cs}$	30.05 (8) years	661.657 (3) <sup>B</sup>	0.8499 (20) <sup>B</sup>	90 years
$^{140}\text{Ba}$ - $^{140}\text{La}$	12.753 (4) days	537.303 (6) 1596.203 (13)	0.2439 (22) 0.9540 (5) <sup>C</sup> 1.1516 (5) <sup>D</sup>	1–1.5 months
$^{144}\text{Ce}$	284.89 (6) days	133.5152 (20)	0.1083 (12)	2–3 years

<sup>A</sup> The lightface numbers in parentheses are the magnitude of plus or minus uncertainties in the last digit(s) listed.

<sup>B</sup> With  $^{137\text{m}}\text{Ba}$  (2.552 min) in equilibrium.

<sup>C</sup> Probability of daughter  $^{140}\text{La}$  decay.

<sup>D</sup> With  $^{140}\text{La}$  (1.67850 d) in transient equilibrium.

<sup>E</sup> Primary reference for half-life, gamma energy, and gamma emission probability is Ref (1) when data is available. Note this reference is to the BIPM data that was recommended at the time of the recommended fission yields were set, that is, as of 2009, and not to the latest Vol 8 data that was published in 2016.

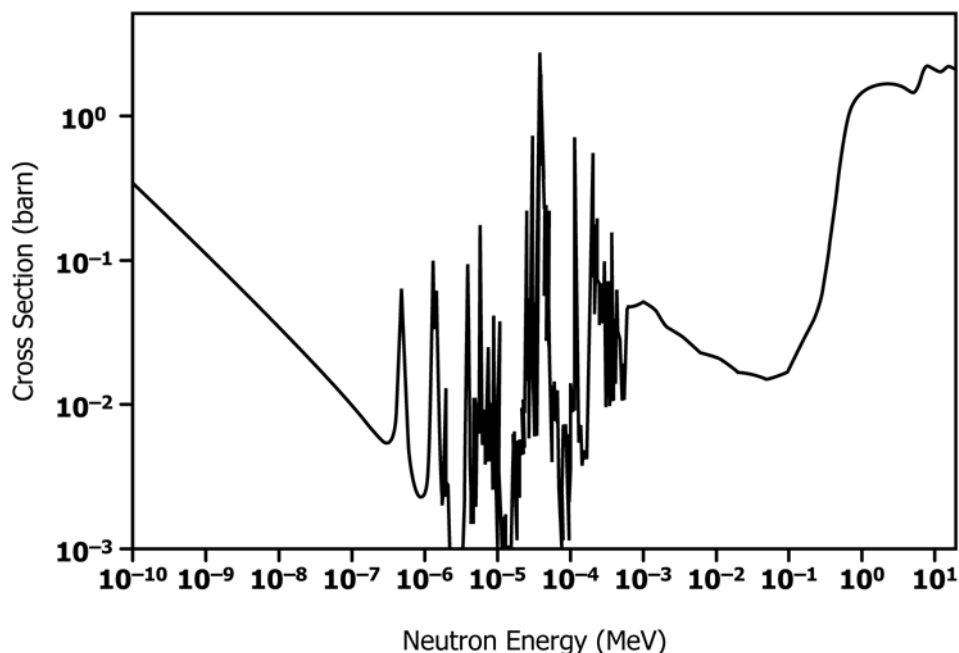


FIG. 1 RRDF/IRDF-1.05 Cross Section Versus Energy for the  $^{237}\text{Np}(n,f)\text{F.P.}$  Reaction

a detector with good sensitivity in the fast neutron region. In the fast fission  $^{252}\text{Cf}$  spontaneous fission benchmark field, ~1 % of the  $^{237}\text{Np}$  fission dosimeter response comes from neutrons with an energy less than 0.1 MeV. In the cavity of a fast burst  $^{235}\text{U}$  reactor, ~5 % of the  $^{237}\text{Np}$  fission dosimeter response comes from neutrons with an energy less than 0.1 MeV. In the cavity of a well-moderated pool-type research reactor ~50 % of the fission response from the  $^{237}\text{Np}(n,f)$  reaction comes from energies less than 0.1 MeV. The importance of this low neutron energy sensitivity should be determined based on the application.

5.6 The  $^{237}\text{Np}$  fission neutron spectrum-averaged cross section in several benchmark neutron fields are given in Table 3 of Practice E261. Sources for the latest recommended cross sections are given in Guide E1018. In the case of the  $^{237}\text{Np}(n,f)\text{F.P.}$  reaction, the recommended cross section source is the Russian Reactor Dosimetry File, RRDF (8). This recommended cross section is identical, for energies up to 20 MeV, to what is found in the latest International Atomic Energy (IAEA) International Reactor Dosimetry and Fusion File, IRDF-1.05 (9). Fig. 1 shows a plot of the recommended cross section versus neutron energy for the fast-neutron reaction  $^{237}\text{Np}(n,f)\text{F.P.}$

## 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 (10):

6.1.1 *Nal(Tl) 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.

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

## 7. Materials

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

7.1.1 The  $^{237}\text{Np}$  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.

NOTE 1—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