



Standard Test Method for Measuring Reaction Rates by Radioactivation of Neptunium- 237¹

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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 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 Dosimetry²
- E 181 Test Methods for Detector Calibration and Analysis of Radionuclides²
- E 261 Practice for Determining Neutron Fluence Rate, Fluence, and Spectra by Radioactivation Techniques²
- E 262 Test Method for Determining Thermal Neutron Reaction and Fluence Rates by Radioactivation Techniques²
- E 320 Test Methods for Cesium-137 in Nuclear Fuel Solutions by Radiochemical Analysis²
- E 393 Test Method for Measuring Reaction Rates by Analysis of Barium-140 from Fission Dosimeters²
- 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-238²
- E 844 Guide for Sensor Set Design and Irradiation for Reactor Surveillance, E706 (IIC)²
- E 944 Guide for Application of Neutron Spectrum Adjust-

ment 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 $^{237}\text{Np}(n,f)\text{F.P.}$

4.2 Various fission products such as ^{137}Cs -, $^{137\text{m}}\text{Ba}$, ^{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.

5.3.1 ^{137}Cs -, $^{137\text{m}}\text{Ba}$ is chosen frequently for long irradiations. Radioactive products ^{134}Cs and ^{136}Cs may be present,

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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 |

^AThe lightface numbers in parentheses are the magnitude of plus or minus uncertainties in the last digit(s) listed.

^BWith ^{137m}Ba (2.552 min) in equilibrium.

^CProbability of daughter ¹⁴⁰La decay.

^DWith ¹⁴⁰La (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 % |

^AEngland, 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.

^BAll yield data given as a %; RC represents a cumulative yield; RI represents an independent yield.

which can interfere with the counting of the 0.662 MeV ¹³⁷Cs–^{137m}Ba gamma ray (see Test Methods E 320).

5.3.2 ¹⁴⁰Ba–¹⁴⁰La is chosen frequently for short irradiations (see Test Method E 393).

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

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

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

5.4.1 Fission product production in a light-water reactor by neutron activation products ²³⁸Np and ²³⁸Pu has been calculated to be insignificant (1.2 %), compared to that from ²³⁷Np(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 ²³⁷Np 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 ²³⁷Np fission and the ⁵⁴Fe(n,p)⁵⁴Mn reaction has been demonstrated (2). The reaction ²³⁷Np(n,f) F.P. is useful since it is responsive to a broader range of neutron energies than most threshold detectors.

5.6 The ²³⁷Np 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 ²³⁷Np(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 ²³⁷Np(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 *Nal(Tl) 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 ²³⁷Np in the form of alloy wire, foil, or oxide powder is available.

7.1.1 The ²³⁷Np 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.

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