

Standard Test Method for Gamma Energy Emission from Fission and Decay Products in Uranium Hexafluoride and Uranyl Nitrate Solution¹

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1. Scope

1.1 This test method covers the measurement of gamma energy emitted from fission products in uranium hexafluoride (UF_6) and uranyl nitrate solution. This test method may also be used to measure the concentration of some uranium decay products. It is intended to provide a method for demonstrating compliance with UF₆ specifications C787 and C996, uranyl nitrate specification C788, and uranium ore concentrate specification C967.

1.2 The lower limit of detection is 5000 MeV Bq/kg (MeV/kg per second) of uranium and is the square root of the sum of the squares of the individual reporting limits of the nuclides to be measured. The limit of detection was determined on a pure, aged natural uranium (ANU) solution. The value is dependent upon detector efficiency and background.

1.3 The fission product nuclides to be measured are ¹⁰⁶Ru/ ¹⁰⁶Rh, ¹⁰³Ru, ¹³⁷Cs, ¹⁴⁴Ce, ¹⁴⁴Pr, ¹⁴¹Ce, ⁹⁵Zr, ⁹⁵Nb, and ¹²⁵Sb. Among the uranium decay product nuclides that may be measured is ²³¹Pa. Other gamma energy-emitting fission and uranium decay nuclides present in the spectrum at detectable levels should be identified and quantified as required by the data quality objectives.

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

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:²
- C761 Test Methods for Chemical, Mass Spectrometric, Spectrochemical, Nuclear, and Radiochemical Analysis of Uranium Hexafluoride
- C787 Specification for Uranium Hexafluoride for Enrichment
- C788 Specification for Nuclear-Grade Uranyl Nitrate Solution or Crystals
- C859 Terminology Relating to Nuclear Materials
- C967 Specification for Uranium Ore Concentrate
- C996 Specification for Uranium Hexafluoride Enriched to Less Than 5 $\%^{235}$ U
- C1022 Test Methods for Chemical and Atomic Absorption Analysis of Uranium-Ore Concentrate
- D3649 Practice for High-Resolution Gamma-Ray Spectrometry of Water
- E181 Test Methods for Detector Calibration and Analysis of Radionuclides

3. Terminology

3.1 Except as otherwise defined herein, definitions of terms are as given in Terminology C859.

4. Summary of Test Method

4.1 A solution of the uranium sample is counted on a high-resolution gamma-ray spectrometry system. The resulting spectrum is analyzed to determine the identity and activity of the gamma-ray-emitting radioactive fission and decay products. The number of counts recorded from one or more of the peaks identified with each fission nuclide is converted to disintegrations of that nuclide per second (Bq). The gamma-ray energy for a fission nuclide is calculated by multiplying the number of disintegrations per second of the nuclide by the

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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.

mean gamma-ray energy emission rate of the nuclide. The calculated gamma-ray energy emission rates for all observed fission nuclides are summed, then divided by the mass of the uranium in the sample to calculate the overall rate of gamma energy production in units of million electron volts per second per kilogram of uranium. Decay product nuclides such as ²³¹Pa will be separately quantified and reported based on specific needs.

5. Significance and Use

5.1 Specific gamma-ray emitting radionuclides in UF₆ are identified and quantified using a high-resolution gamma-ray energy analysis system, which includes a high-resolution germanium detector. This test method shall be used to meet the health and safety specifications of C787, C788, and C996 regarding applicable fission products in reprocessed uranium solutions. This test method may also be used to provide information to parties such as conversion facilities on the level of uranium decay products in such materials. Pa-231 is a specific uranium decay product that may be present in uranium ore concentrate and is amenable to analysis by gamma spectrometry.

6. Apparatus

6.1 *High-Resolution Gamma-Ray Spectrometry System*, as specified in Practice D3649. The energy response range of the spectrometry system may need to be tailored to address all the needed fission and uranium decay product nuclides that need to be analyzed for.

6.2 Sample Container with Fitted Cap—A leak-proof plastic container capable of holding the required sample volume. The dimensions must be consistent between containers used for samples and standard to keep the counting geometry constant. The greatest detection efficiency will be achieved with a low-height sample container with a diameter slightly smaller than the detector being used.

6.3 *Sample Holder*, shall be used to position the sample container such that the detector view of the sample is reproducible. To reduce the effects of coincident summing, the sample holder shall provide a minimum separation of 5 mm between the sample container and the detector end cap.

7. Calibration and Standardization of Detector

7.1 Prepare a mixed radionuclide calibration standard stock solution covering the energy range of approximately 50 to 2000 keV.

7.1.1 Commercial calibration standards are available which are traceable to NIST or other national standards laboratories.

7.2 Prepare a solution of ANU at 6.74 gU/100 g. The uranium and its progeny's relationship must not have been altered for at least eight months.

7.3 Transfer a known, suitable activity of the mixed nuclide calibration standard stock solution (40 to 50 kBq) to a container identical to that used for the sample measurement. Add ANU solution to the mixed nuclide standard so that the final volume and uranium concentration match those expected in the sample measurement. Test Methods E181 and Practice D3649 provides information on calibration of detector energy, efficiency, resolution, and other parameters.

7.4 The detector energy scale and efficiency are calibrated by placing the container with the mixed nuclide calibration standard in a sample holder that provides a reproducible geometry relative to the detector. Collect a spectrum over a period up to 1 h that includes all the gamma photopeaks in the energy range up to ~ 2000 keV. All counting conditions (except count duration) must be identical to those that will be used for analysis of the actual sample.

7.5 Determine the net counts under each peak of every nuclide in the mixed radionuclide standard, then divide by the count duration (live time) to determine the rate in counts per second for each radionuclide. If a background count on the detector shows any net peak area for the peaks of interest, these must be subtracted from the standard counts per second.

7.6 Divide the observed count rate determined for each gamma peak by the calculated emission rate of the gamma ray that produced the peak in the mixed calibration standard (gammas per second).

7.6.1 Calculation of the gamma emission rate for each peak from the mixed calibration standard must account for the following:

Nuclide	Half- Life	Decay Constant (λ ₁)	Measurement Peaks, MeV	Abundance Gamma/ Disintegration (G _I)	Mean Gamma Energy Disintegration, MeV Bq (E ₁)
¹⁰³ Ru/ ¹⁰³ Rh	39.35d	0.01761/d	0.4971	0.889	0.484
			0.6103	0.056	
¹⁰⁶ Ru/ ¹⁰⁶ Rh	366.5d	0.001891/d	0.5119	0.207	0.209
			0.6222	0.0981	
¹⁴¹ Ce	32.55d	0.02129/d	0.1454	0.484	0.0718
¹⁴⁴ Ce/ ¹⁴⁴ Pr	284.5d	0.002436/d	0.1335	0.1110	0.0518
¹³⁷ Cs/ ¹³⁷ Ba	30.17y	0.02297/y	0.6616	0.851	0.5655
⁹⁵ Nb	34.97d	0.01982/d	0.7658	1.000	0.766
⁹⁵ Zr	63.98d	0.01083/d	0.7242	0.444	0.737
			0.7567	0.549	
¹²⁵ Sb	2.71y	0.256/y	0.4279	0.294	0.433
			0.6008	0.178	
²³¹ Pa	32760y	2.1158E-05/y	0.002736	0.103	n/a

TABLE 1 Gamma-Ray-Emitting Fission and Decay Products Found in UF₆

7.6.1.1 Activity of the nuclide that produces the peak in its original standard (disintegrations/second/unit volume). This is taken from the standard certificate of measurement supplied with the standard.

7.6.1.2 Volume of each isotopic standard taken for the mixed standard and the final volume of the mixed standard.

7.6.1.3 Fraction of the volume of the mixed standard taken for counting.

7.6.1.4 Decay of the activity of each isotope in the standard between its date of standardization and the date of counting according to the equation:

$$A_i = A_{i_0} e^{-\lambda_i t} \tag{1}$$

where:

 A_i = activity of isotope *i* on the date of counting in Bq,

- A_{i_0} = activity of isotope *i* on the date of standard characterization in Bq,
- λ_i = decay constant of isotope *i* in units of inverse time (values for some isotopes of interest may be found in column 3 of Table 1), and
- *t* = elapsed time between the calibration reference date and the date of counting. Time units must be the same as in the decay constant.

7.6.1.5 The abundance of gamma rays of the energy of interest emitted by each disintegration (see Table 1).

7.7 Plot a detector efficiency curve of counts/gamma versus gamma energy. Most multichannel analyzers and associated software are able to store individual values from this curve or the equation of the curve for later use.

7.8 This efficiency calibration will remain valid provided none of the sample or instrument parameters are changed (for example, volume of sample, container geometry, distance from detector, and detector) and instrument response to the control standard remains within the statistical limits established.

8. Measurement of Control Standard Solution

8.1 Measure the control standard solution prepared in 7.3 with the geometry as used during detector efficiency calibration. Ten measurements of the control standard solution are made. The calculated data for the fission products is used to establish precision and bias of the test method.

8.1.1 Most multichannel analyzers and associated software have automatic routines for determining the net counts under single peaks and double peaks that are not resolved. If the available analyzer does not have such capabilities, refer to Reilly³ for single-peak analysis methods and 8.2.1 and 8.2.2 for double-peak problems that are likely to be encountered.

8.1.2 Peaks that are determined for this analysis are listed in Table $1,^4$ along with the abundance factors, decay constants,

and the mean gamma energy per disintegration for each nuclide. Needed information for uranium decay products can be found in Footnote 4^4 or other available sources.

8.2 While most full-energy gamma emissions are generally characteristic of specific radionuclides, it is possible that unresolved multiplets may produce biased peak areas. Determination of the following peak areas may cause problems during calibration or sample measurements.

8.2.1 The peak produced by the 765.9-keV gamma ray of 95 Nb is not resolved from the peak produced by the 766.4-keV gamma ray of 234m Pa, a progeny radionuclide of 238 U. The following procedure is suggested to determine the count rate of 95 Nb in the double peak.

8.2.1.1 Perform a series of count measurements for periods up to 1 h of a sample of ANU under the same conditions as the calibration standard or sample. The counting period should be adjusted so that the counting uncertainties are less than 1 % for the appropriate peaks of interest.

8.2.1.2 For each measurement, determine the ratio of counts in the 234m Pa peaks at 766.4 and 1001 keV using the equation:

$$R_{\rm Pa} = C_{766 \text{ total}} / C_{1001} \tag{2}$$

where:

$$R_{\text{Pa}}$$
 = ratio of counts in the 766.4 and 1001-keV peaks of ^{234m}Pa,

 $C_{766 \text{ total}} = \text{total counts in the double peak near 766 keV, and} C_{1001} = \text{counts in the 1001-keV peak of}^{234\text{m}}\text{Pa.}$

8.2.1.3 Calculate the mean value for the ratio (\bar{R}_{Pa}).

8.2.1.4 Determine the 95 Nb counts at 765.9 keV by use of the equation:

$$C_{\rm Nb} = C_{766 \text{ total}} - \left[(C_{1001}) (\overline{R}_{\rm Pa}) \right]$$
 (3)

where:

 $C_{\rm Nb}$ = counts in the peak near 766 keV resulting from 765.9-keV gamma rays of ⁹⁵Nb.

8.2.2 The peak produced by the 145.4-keV gamma ray of 141 Ce is not resolved from the peak produced by the 143.8-keV gamma ray of 235 U. The following procedure is suggested to determine the count rate of 141 Ce in the double peak.

8.2.2.1 Perform a series of measurements of up to 1-h counting time of a sample of ANU under the same conditions as the calibration standard or sample.

8.2.2.2 For each measurement, determine the ratio of counts in the 235 U peaks at 143.8 and 185.7 keV using the equation:

$$R_{\rm U} = C_{144 \text{ total}} / C_{185.7} \tag{4}$$

where:

 $R_{\rm U}$ = ratio of counts in the 143.8 and 185.7-keV peaks of ²³⁵U,

 $C_{144 \text{ total}} = \text{total counts in the double peak near 144 keV, and} C_{185.7} = \text{counts in the 185.7-keV peak of }^{235}\text{U.}$

8.2.2.3 Calculate the mean value for the ratio (\bar{R}_{II}).

8.2.2.4 Determine the 141 Ce counts at 145.4 keV by use of the equation:

$$C_{\rm Ce} = C_{\rm 144\ total} - \left[\left(C_{\rm 185} \right) \left(\overline{R}_{\rm U} \right) \right] \tag{5}$$

³ Reilly, T. D., and Parker, J. L., *A Guide to Gamma-Ray Assay for Nuclear Materials Accountability*, LA-5794-M, Los Alamos National Laboratory, 1975. DOI: 10.2172/4210151.

⁴ The information in Table 1 for fission products is from the Joint European File: 1 data file supplied by the Nuclear Energy Agency, Paris, France. The user may use other published data. The uranium decay product information in Table 1 is from L.P. Ekström and R.B. Firestone, WWW Table of Radioactive Isotopes, database version 2/28/99 from URL http://ie.lbl.gov/toi/index.htm. The user may use other published data for uranium decay products.

where:

 C_{Ce} = counts in the peak near 144 keV resulting from 145.4-keV gamma rays of ¹⁴¹Ce.

9. Procedure

9.1 Hydrolyze a UF_6 sample as in Test Method C761, dissolve a uranium ore concentrate sample using suitable approach in Test Method C1022, or prepare the uranyl nitrate solution sample. Ensure that sample preparation parameters (solution volume, uranium concentration, sample container, geometry, and so forth) are the same as used during detector efficiency calibration. Note the mass of uranium (*W*) taken in grams.

9.2 Place the container and sample into the counter with the same geometry as used during detector efficiency calibration. Count the sample for 60 min to collect a gamma spectrum of the sample.

9.3 Determine the net counts under one or more peaks for each nuclide, then divide by the count duration (live time) to determine the count rate for each gamma peak in counts per second. See 8.2.1 and 8.2.2 for methods to deal with unresolved double peaks.

10. Calculation

10.1 Determine the gamma energy release rate for each nuclide according to the following equation:

$$F_i = \frac{1000}{W} \times \frac{C_i}{Eff \times G_i} \times E_i \tag{6}$$

where:

- F_i = rate of energy released in gamma radiation as a result of fission nuclide *i* decay in MeV Bq/kg U (MeV/kg per second),
- C_i = count rate calculated in 9.3 for a single gamma-ray peak of nuclide *i* (counts per second),
- *Eff* = the detector efficiency (counts/gamma) determined in Section 7 for the energy of the gamma-ray peak being analyzed,
- G_i = the gamma-ray production rate (gammas/ disintegration) by nuclide *i* for the energy of gamma ray being analyzed (from Table 1),
- E_i = mean gamma energy release per disintegration of nuclide *i* in MeV (from Table 1), and
- W = uranium sample weight, in grams.

10.2 Determine the total fission product energy release rate, F_{Total} , by summing the contributions from all nuclides detected, as follows (expressed in units of MeV Bq/kg U (MeV/kg U per second)).

$$F_{\text{Total}} = \sum F_i \tag{7}$$

10.3 Uranium decay product nuclide concentrations can be calculated from the detector efficiency curve (7.7), sample quantity information, and gamma abundance and other nuclide data from Table 1, Footnote 4, or other sources. Uranium decay product gamma energy shall not be included in the total fission product energy release rate calculation in Eq 7.

11. Precision and Bias

11.1 Within the different stages of the nuclear fuel cycle many challenges lead to the inability to perform interlaboratory studies for precision and bias. These challenges may include variability of matrices of material tested, lack of suitable reference or calibration materials, limited laboratories performing testing, shipment of materials to be tested, and regulatory constraints. Because of these challenges each laboratory utilizing these test methods should develop their own precision and bias as part of their quality assurance program.

11.2 Precision:

11.2.1 Precision data was obtained from ten measurements of a uranyl fluoride (UO_2F_2) solution prepared from ANU hexafluoride and spiked nuclides ¹⁰⁶Ru, ¹³⁴Cs, ⁶⁰Co, and ¹³⁷Cs from an international traceable standard. The work was carried out by one analyst over a period of weeks, and the data is in Table 2.

11.3 Bias Estimate:

11.3.1 No standard material is certified for fission products in UF_6 or UNO_2 solution. The bias estimates were obtained from the same data used to calculate the precision. The data are summarized in Table 2.

11.3.2 The data gave a relative bias of -18 % for 106 Ru and -12 % for 134 Cs. This negative bias is probably because of the effects of coincidence summing and absorption.

12. Keywords

12.1 decay products; fission products; high purity germanium detector; gamma energy; gamma-ray spectrometry; uranium hexafluoride; uranium nitrate

TABLE 2 Precision and Bias Data

Nuclide	Prepared	Measured	Standard Deviation of
	Activity Level,	Activity Level,	Measured Activity (1s),
	MeV Bq/kg U	MeV Bq/kg U	MeV Bq/kg U
¹⁰⁶ Ru	1.7×10^{5}	1.4×10^{5}	1.10×10^{3}
¹³⁴ Cs	1.0×10^{5}	8.8×10^{4}	1.40×10^{4}
⁶⁰ Co	1.2×10^{5}	1.2×10^{5}	1.79×10^{3}
¹³⁷ Cs	2.4×10^{4}	2.4×10^{4}	3.40×10^{2}

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