



# Standard Test Method for Gamma Energy Emission from Fission Products in Uranium Hexafluoride and Uranyl Nitrate Solution<sup>1</sup>

This standard is issued under the fixed designation C 1295; 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 the measurement of gamma energy emitted from fission products in uranium hexafluoride ( $\text{UF}_6$ ) and uranyl nitrate solution. It is intended to provide a method for demonstrating compliance with  $\text{UF}_6$  specifications C 787 and C 996 and uranyl nitrate specification C 788.

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 nuclides to be measured are  $^{106}\text{Ru}/^{106}\text{Rh}$ ,  $^{103}\text{Ru}$ ,  $^{137}\text{Cs}$ ,  $^{144}\text{Ce}$ ,  $^{144}\text{Pr}$ ,  $^{141}\text{Ce}$ ,  $^{95}\text{Zr}$ ,  $^{95}\text{Nb}$ , and  $^{125}\text{Sb}$ . Other gamma energy-emitting fission nuclides present in the spectrum at detectable levels should be identified and quantified as required by the data quality objectives.

1.4 *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:*<sup>2</sup>

C 761 Test Methods for Chemical, Mass Spectrometric, Spectrochemical, Nuclear, and Radiochemical Analysis of Uranium Hexafluoride

C 787 Specification for Uranium Hexafluoride for Enrichment

C 788 Specification for Nuclear-Grade Uranyl Nitrate Solution

C 996 Specification for Uranium Hexafluoride Enriched to Less than 5 %<sup>235</sup>U

<sup>1</sup> This test method is under the jurisdiction of ASTM Committee C26 on Nuclear Fuel Cycle and is the direct responsibility of Subcommittee C26.05 on Methods of Test.

Current edition approved July 1, 2005. Published August 2005. Originally approved in 1995. Last previous edition approved in 1998 as C 1295 – 98.

<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.

D 3649 Practice for High Resolution Gamma-Ray Spectrometry of Water

## 3. Summary of Test Method

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

## 4. Significance and Use

4.1 The gamma-ray emitting fission products in  $\text{UF}_6$  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 C 787, C 788, and C 996 regarding applicable fission products in reprocessed uranium solutions.

## 5. Apparatus

5.1 *High-Resolution Gamma-Ray Spectrometry System*, as specified in Practice D 3649.

5.2 *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.

5.3 *Sample Holder*, shall be used to position the sample container such that the detector view of the sample is reproducible. To minimize 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.

## 6. Calibration and Standardization of Detector

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

6.1.1 Commercial calibration standards are available.

6.2 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.

6.3 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. Practice D 3649 provides information on calibration of detector energy, efficiency, resolution, and other parameters.

6.4 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 time) must be identical to those that will be used for analysis of the actual sample.

6.5 Determine the net counts under each peak of every nuclide in the mixed radionuclide standard, then divide by the count time (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.

6.6 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).

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

6.6.1.1 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.

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

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

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

$$A_i = A_{i_0} e^{-\lambda_i t} \quad (1)$$

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,

$\lambda_i$  = 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.

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

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

6.8 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.

## 7. Measurement of Control Standard Solution

7.1 Measure the control standard solution prepared in 6.3 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.

7.1.1 Most multichannel analyzers 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 Reilly<sup>3</sup> for single-peak analysis methods and 7.2.1 and 7.2.2 for double-peak problems that are likely to be encountered.

7.1.2 Peaks that are determined for this analysis are listed in Table 1,<sup>4</sup> along with the abundance factors, decay constants, and the mean gamma energy per disintegration for each nuclide.

7.2 Determination of the following peak areas may cause problems during calibration or sample measurements.

7.2.1 The peak produced by the 765.9-keV gamma ray of <sup>95</sup>Nb is not resolved from the peak produced by the 766.4-keV gamma ray of <sup>234m</sup>Pa, a daughter of <sup>238</sup>U. The

TABLE 1 Gamma-Ray-Emitting Fission Products Found in UF<sub>6</sub>

Nuclide	Half-Life	Decay Constant ( $\lambda_i$ )	Measurement Peaks, MeV	Abundance Gamma/Disintegration ( $G_i$ )	Mean Gamma Energy Disintegration, MeV Bq ( $E_i$ )
<sup>103</sup> Ru/ <sup>103</sup> Rh	39.35d	0.01761/d	0.4971	0.889	0.484
			0.6103	0.056	
<sup>106</sup> Ru/ <sup>106</sup> Rh	366.5d	0.001891/d	0.5119	0.207	0.209
			0.6222	0.0981	
<sup>141</sup> Ce	32.55d	0.02129/d	0.1454	0.484	0.0718
<sup>144</sup> Ce/ <sup>144</sup> Pr	284.5d	0.002436/d	0.1335	0.1110	0.0518
<sup>137</sup> Cs/ <sup>137</sup> Ba	30.17y	0.02297/y	0.6616	0.851	0.5655
<sup>95</sup> Nb	34.97d	0.01982/d	0.7658	1.000	0.766
<sup>95</sup> Zr	63.98d	0.01083/d	0.7242	0.444	0.737
			0.7567	0.549	
<sup>125</sup> Sb	2.71y	0.256/y	0.4279	0.294	0.433
			0.6008	0.178	

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

<sup>4</sup> The information in Table 1 is from the Joint European File: 1 data file supplied by the Nuclear Energy Agency, Paris, France. The user may use other published data.

following procedure is suggested to determine the count rate of  $^{95}\text{Nb}$  in the double peak.

7.2.1.1 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 errors are less than 1 % for the appropriate peaks of interest.

7.2.1.2 For each measurement, determine the ratio of counts in the  $^{234\text{m}}\text{Pa}$  peaks at 766.4 and 1001 keV using the equation:

$$R_{\text{Pa}} = C_{766 \text{ total}} / C_{1001} \quad (2)$$

where:

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

$C_{766 \text{ total}}$  = total counts in the double peak near 766 keV, and

$C_{1001}$  = counts in the 1001-keV peak of  $^{234\text{m}}\text{Pa}$ .

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

7.2.1.4 Determine the  $^{95}\text{Nb}$  counts at 765.9 keV by use of the equation:

$$C_{\text{Nb}} = C_{766 \text{ total}} - [(C_{1001})(\bar{R}_{\text{Pa}})] \quad (3)$$

where:

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

7.2.2 The peak produced by the 145.4-keV gamma ray of  $^{141}\text{Ce}$  is not resolved from the peak produced by the 143.8-keV gamma ray of  $^{235}\text{U}$ . The following procedure is suggested to determine the count rate of  $^{141}\text{Ce}$  in the double peak.

7.2.2.1 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.

7.2.2.2 For each measurement, determine the ratio of counts in the  $^{235}\text{U}$  peaks at 143.8 and 185.7 keV using the equation:

$$R_{\text{U}} = C_{144 \text{ total}} / C_{185.7} \quad (4)$$

where:

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

$C_{144 \text{ total}}$  = total counts in the double peak near 144 keV, and

$C_{185.7}$  = counts in the 185.7-keV peak of  $^{235}\text{U}$ .

7.2.2.3 Calculate the mean value for the ratio ( $\bar{R}_{\text{U}}$ ).

7.2.2.4 Determine the  $^{141}\text{Ce}$  counts at 145.4 keV by use of the equation:

$$C_{\text{Ce}} = C_{144 \text{ total}} - [(C_{185})(\bar{R}_{\text{U}})] \quad (5)$$

where:

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

## 8. Procedure

8.1 Hydrolyze the  $\text{UF}_6$  sample for counting as in Test Method C 761 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.

8.2 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.

8.3 Determine the net counts under one or more peaks for each nuclide, then divide by the count time (live time) to determine the count rate for each gamma peak in counts per second. See 7.2.1 and 7.2.2 for methods to deal with unresolved double peaks.

## 9. Calculation

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

$$F_i = \frac{1000}{W} \times \frac{C_i}{\text{Eff} \times G_i} \times E_i \quad (6)$$

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 8.3 for a single gamma-ray peak of nuclide  $i$  (counts per second),

$\text{Eff}$  = the detector efficiency (counts/gamma) determined in Section 6 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.

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

$$F_{\text{Total}} = \sum F_i \quad (7)$$

## 10. Precision and Bias

### 10.1 Precision:

10.1.1 Precision data was obtained from ten measurements of a uranyl fluoride ( $\text{UO}_2\text{F}_2$ ) solution prepared from ANU hexafluoride and spiked nuclides  $^{106}\text{Ru}$ ,  $^{134}\text{Cs}$ ,  $^{60}\text{Co}$ , and  $^{137}\text{Cs}$  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.

### 10.2 Bias Estimate:

10.2.1 No standard material is certified for fission products in  $\text{UF}_6$  or  $\text{UNO}_2$  solution. The bias estimates were obtained

TABLE 2 Precision and Bias Data

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

from the same data used to calculate the precision. The data are summarized in **Table 2**.

10.2.2 The data gave a relative bias of  $-18\%$  for  $^{106}\text{Ru}$  and  $-12\%$  for  $^{134}\text{Cs}$ . This negative bias is probably because of the effects of coincidence summing and absorption.

## 11. Keywords

11.1 fission products; gamma energy; uranium hexafluoride; uranium nitrate

*ASTM International takes no position respecting the validity of any patent rights asserted in connection with any item mentioned in this standard. Users of this standard are expressly advised that determination of the validity of any such patent rights, and the risk of infringement of such rights, are entirely their own responsibility.*

*This standard is subject to revision at any time by the responsible technical committee and must be reviewed every five years and if not revised, either reapproved or withdrawn. Your comments are invited either for revision of this standard or for additional standards and should be addressed to ASTM International Headquarters. Your comments will receive careful consideration at a meeting of the responsible technical committee, which you may attend. If you feel that your comments have not received a fair hearing you should make your views known to the ASTM Committee on Standards, at the address shown below.*

*This standard is copyrighted by ASTM International, 100 Barr Harbor Drive, PO Box C700, West Conshohocken, PA 19428-2959, United States. Individual reprints (single or multiple copies) of this standard may be obtained by contacting ASTM at the above address or at 610-832-9585 (phone), 610-832-9555 (fax), or [service@astm.org](mailto:service@astm.org) (e-mail); or through the ASTM website ([www.astm.org](http://www.astm.org)).*