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Standard Test Method for Gamma Energy Emission from Fission Products in Uranium Hexafluoride and Uranyl Nitrate Solution[1]

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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. It is intended to provide a method for demonstrating compliance with UF_6 Specifications C 787 and C 996 and replace the ion chamber measurement technique.

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 $^{106}Ru/^{106}Rh$, ^{103}Ru, ^{144}Ce, ^{144}Pr, ^{141}Ce, ^{95}Zr, ^{95}Nb, and ^{125}Sb. Other gamma energy-emitting fission nuclides that are identified should be measured and reported.

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:*

C 761 Test Methods for Chemical, Mass Spectrometric, Spectrochemical, Nuclear, and Radiochemical Analysis of Uranium Hexafluoride[2]

C 787 Specification for Uranium Hexafluoride for Enrichment[2]

C 996 Specification for Uranium Hexafluoride Enriched to Less than 5 % ^{235}U[2]

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 fission products in UF_6 are identified and quantified using a high-resolution gamma-ray energy analysis system, which includes a high-resolution germanium detector.

5. Apparatus

5.1 *High-Resolution Germanium Detector*.

5.1.1 A relative efficiency of 10 % or higher (ratio of area under the 1.33-MeV photopeak of Cobalt-60 to that obtained with a 76- by 76-mm (3- by 3-in.) NaI(T1) detector, at a source-to-detector distance of 25 cm).

5.1.2 Full width at half maximum (FWHM) resolution of 2.3 keV or less at 1.33 MeV.

5.2 *Gamma-Ray Analysis System*, containing the following components:

5.2.1 High voltage bias power supply.

5.2.2 Spectroscopy amplifier with peak shaping time constants of 1, 2 and 4 μs, baseline restoration, pulse pileup rejection, and live time correction.

5.2.3 Multichannel analyzer with a minimum of 4096 data channels and circuits compatible with the pulse pileup rejection and live time correction circuits of the spectroscopy amplifier.

5.2.4 Computer system and input/output (I/O) devices for storage of calibration data, control of analyzer functions, calculation of results, and report generation.

5.3 *Heavy Metal Shield*, for the detector and sample.

5.4 *Sample Container Fitted with Cap*, internal diameter 56 mm, outside diameter 59 mm, and height 65 mm. The dimensions must be consistent between sample containers to keep the counting geometry constant.

5.5 *Sample Holder*, shall accommodate the sample container and position it so that the detector view of the sample container is reproducible. To minimize the effects of coincident

[1] This test method is under the jurisdiction of ASTM Committee C-26 on Nuclear Fuel Cycle and is the direct responsibility of Subcommittee C26.05 on Methods of Test.

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[2] *Annual Book of ASTM Standards*, Vol 12.01.

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[3] covering the energy range of 5 to 2000 keV.

6.2 Prepare a solution of ANU at 6.74 gU/100 g. The uranium and its daughter products' 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. This test method is used to calibrate the energy scale of the detector and determine the counter efficiency.

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 Volume of the mixed standard taken for counting.

6.6.1.4 Decay of the activity of the standard between its date of characterization and the date of counting according to the equation:

$$A = A_O e^{-\lambda t} \quad (1)$$

where:

A = activity on the date of counting disintegration/s;

A_0 = activity on the date of standard characterization in disintegration/s;

λ = decay constant in units of time^{-1}, values from Table 1; and

t = time between calibration reference date and the date of counting. Time units must be the same as 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. Some multichannel analyzers and data reduction programs may be 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 Many 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[4] 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,[5] 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 be expected to cause problems during both calibration and sample measurements.

[3] The mixed radionuclide calibration standard QCY48 from Amersham International, White Lion Road, Amersham, Buckinghamshire, HP7 9LL, England, has been found to be suitable.

[4] Reilly, T. D., and Parker, J. L., *A Guide to Gamma Ray Assay for Nuclear Materials Accountability*, LA-5794M, Los Alamos National Laboratory, 1975.

[5] 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.

TABLE 1 Gamma-Ray-Emitting Fission Products Found in UF_6

Nuclide	Half-Life	Decay Constant	Measurement Peaks, MeV	Abundance Gamma/Disintegration	Mean Gamma Energy Disintegration, MeV
$^{103}Ru/^{103}Rh$	39.35d	0.01761/d	0.4971	0.889	0.497
			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	0.0765
$^{144}Ce/^{144}Pr$	284.5d	0.002436/d	0.1335	0.1110	0.0479
^{134}Cs	2.062y	0.3362/y	0.7958	0.851	1.554
$^{137}Cs/^{137}Ba$	30.17y	0.02297/y	0.6616	0.899	0.597
^{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	

7.2.1 The peak produced by the 765.9-keV gamma ray of $_{95}$Nb is not resolved from the peak produced by the 765-keV gamma ray of ^{234m}Pa, a daughter of ^{238}U. The following procedure is suggested to determine the count rate of ^{95}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 of the ^{234m}Pa peaks at 765 and 1001 keV, then calculate the mean value for the ratio.

$$R_{Pa} = (\text{counts at 765 keV})/(\text{counts at 1001 keV}) \quad (2)$$

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

$$C_{Nb} = C_{766TOT} - [(C_{1001})(R_{Pa})] \quad (3)$$

where:

C_{Nb} = counts in 765.9-keV peak as a result of ^{95}Nb,
C_{766TOT} = total counts in the double peak near 766 keV, and
C_{1001} = counts in the 1001-keV peak of ^{234m}Pa.

7.2.2 The peak produced by the 145.4-keV gamma ray of ^{141}Ce is not resolved from the peak produced by the 143-keV gamma ray of ^{235}U. The following procedure is suggested to determine the count rate of ^{141}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 of the ^{235}U peaks at 144 and 185 keV, then calculate the mean value for the ratio as follows:

$$R_u = (\text{counts at 143 keV})/(\text{counts at 185 keV}) \quad (4)$$

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

$$C_{Ce} = C_{144TOT} - [(C_{185})(R_u)] \quad (5)$$

where:

C_{Ce} = counts in 145.4-keV peak as a result of ^{141}Ce,
C_{144TOT} = total counts in double peak near 144 keV, and
C_{185} = counts in the 185-keV peak of ^{235}U.

8. Procedure

8.1 Hydrolyze the 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, *W*, of uranium taken (g).

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:

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

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),
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, g.

9.2 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)).

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

10. Precision and Bias

10.1 *Precision*:

10.1.1 Precision data was obtained from ten measurements of a uranyl fluoride (UO_2F_2) solution prepared from ANU hexafluoride and spiked nuclides ^{106}Ru, ^{134}Cs, ^{60}Co, and ^{137}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*:

10.2.1 Accuracy data was obtained from the ten measurements on the same UO_2F_2 solution used for precision. The data is in Table 2.

10.2.2 The data gave a relative bias of −18 % for ^{106}Ru and −12 for ^{134}Cs. These results are probably biased low because of the effects of coincidence summing and absorbtion.

11. Keywords

11.1 fission products; gamma energy; uranium hexafluoride

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}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