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

This standard is issued under the fixed designation C1295; 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 (ε) 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 and decay products in uranium hexafluoride (UF_6) and uranyl nitrate solution. It is intended to provide a method for demonstrating compliance with UF_6 specifications C787 and C996 ~~and~~ , 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~~ are ^{106}Ru/^{106}Rh, ^{103}Ru, ^{137}Cs, ^{144}Ce, ^{144}Pr, ^{141}Ce, ^{95}Zr, ^{95}Nb, and ^{125}Sb. Among the uranium decay product nuclides that may be measured is ^{231}Pa. 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
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

3. Summary of Test Method

3.1 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 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 ^{231}Pa will be separately quantified and reported based on specific needs.

[1] 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.
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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.

4. Significance and Use

4.1 Specific gamma-ray emitting radionuclides in 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 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.

5. Apparatus

5.1 *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.

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 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.

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 which are traceable to NIST or other national standards laboratories.

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 D3649 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 duration) 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 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.

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).

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

Nuclide	Half-Life	Decay Constant (λ_I)	Measurement Peaks, MeV	Abundance Gamma/ Disintegration (G_I)	Mean Gamma Energy Disintegration, MeV Bq (E_I)
$^{103}Ru/^{103}Rh$	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

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,
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.

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~~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.~~

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 associated software 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 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 Reilly[3] 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,[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 Reference 4[4] or other available sources.

7.2 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.

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 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.

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

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

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

[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.