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Standard Test Method for Determination of Uranium Isotopic Composition by Gamma-Ray Spectrometry¹

This standard is issued under the fixed designation C1931; 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 applies to the nondestructive determination of the isotopic abundances of uranium, typically ^{234}U , ^{235}U , ^{236}U , and ^{238}U , in isotopically homogeneous uranium-bearing materials using gamma spectrometry. The material is commonly inside a container and is measured without specimen preparation.

1.2 This test method is applicable to items containing sub-gram quantities of uranium to the maximum uranium mass allowed by criticality considerations.

1.3 Measurable gamma ray emissions from uranium cover the energy range from below 80 keV to above 1000 keV. K-X-ray emissions from the isotopes of uranium and their daughters are found in the energy region around 100 keV. This test method has been applied to all portions of this energy range.

1.4 The isotopic abundance of ^{236}U is usually not directly determined because its low-energy gamma rays are too weak **(1)**² to be detected under normal measurement conditions. Isotopic correlation techniques have been used to estimate its relative abundance **(2)**.

1.5 This test method has been demonstrated in routine use for isotopic amount fraction (atom %) of ^{235}U from 0.2 % to 97 %.

1.6 This test method requires decay equilibrium (160 days for 99 %) between ^{238}U and its 24.1 d half-life ^{234}Th daughter. Corrections can be made if the date of chemical separation of the ^{234}Th daughter is known.

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

1.8 *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, health, and environmental practices and determine the applicability of regulatory limitations prior to use.

1.9 *This international standard was developed in accordance with internationally recognized principles on standardization established in the Decision on Principles for the Development of International Standards, Guides and Recommendations issued by the World Trade Organization Technical Barriers to Trade (TBT) Committee.*

2. Referenced Documents

2.1 ASTM Standards:³

- C698 Test Methods for Chemical, Mass Spectrometric, and Spectrochemical Analysis of Nuclear-Grade Mixed Oxides ((U, Pu)O₂)
- C1030 Test Method for Determination of Plutonium Isotopic Composition by Gamma-Ray Spectrometry
- C1133 Test Method for Nondestructive Assay of Special Nuclear Material in Low-Density Scrap and Waste by Segmented Passive Gamma-Ray Scanning
- C1221 Test Method for Nondestructive Analysis of Special Nuclear Materials in Homogeneous Solutions by Gamma-Ray Spectrometry
- C1316 Test Method for Nondestructive Assay of Nuclear Material in Scrap and Waste by Passive-Active Neutron Counting Using ^{252}Cf Shuffler
- C1455 Test Method for Nondestructive Assay of Special Nuclear Material Holdup Using Gamma-Ray Spectroscopic Methods
- C1458 Test Method for Nondestructive Assay of Plutonium, Tritium and ^{241}Am by Calorimetric Assay
- C1493 Test Method for Non-Destructive Assay of Nuclear Material in Waste by Passive and Active Neutron Counting Using a Differential Die-Away System
- C1514 Test Method for Measurement of ^{235}U Fraction Using Enrichment Meter Principle
- C1673 Terminology of C26.10 Nondestructive Assay Methods

¹ This test method is under the jurisdiction of ASTM Committee C26 on Nuclear Fuel Cycle and is the direct responsibility of Subcommittee C26.10 on Non Destructive Assay.

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² The boldface numbers in parentheses refer to the list of references at the end of this standard.

³ 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.

C1718 Test Method for Nondestructive Assay of Radioactive Material by Tomographic Gamma Scanning

E181 Guide for Detector Calibration and Analysis of Radionuclides in Radiation Metrology for Reactor Dosimetry

E267 Test Method for Uranium and Plutonium Concentrations and Isotopic Abundances

2.2 *ANSI Standards*:⁴

ANSI/IEEE Std 325-1996 IEEE Standard Test Procedures for Germanium Gamma-Ray Detectors

ANSI N15.36 Measurement Control Program – Nondestructive Assay Measurement Control and Assurance

3. Summary of Test Method

3.1 The full-energy peak intensities of gamma-rays emitted from an uranium-bearing item are determined from a single gamma-ray spectrum obtained with a High-Purity Germanium (HPGe) detector. The method has also been developed for use with LaBr₃(Ce) and CZT detectors (3).

3.2 For isotopically homogeneous uranium the isotope amount ratio, N^i/N^k , for isotopes i and k is related to the net counts in the full energy peaks of interest, $C(E_j^i)$, for gamma ray j with energy E_j emitted from isotope i and $C(E_l^k)$ for gamma ray l with energy E_l from isotope k by:

$$\frac{N^i}{N^k} = \frac{C(E_j^i)}{C(E_l^k)} \cdot \frac{T_{1/2}^i}{T_{1/2}^k} \cdot \frac{BR_j^k}{BR_l^i} \cdot \frac{RE(E_l)}{RE(E_j)} \quad (1)$$

where:

$RE(E_i)$ = relative detection efficiency for a gamma-ray of energy E_i ,

$T_{1/2}^i$ = half-life of isotope i , and

BR_j^i = gamma-ray branching ratio or branching intensity (usually expressed as gamma-rays per disintegration) of gamma ray j from isotope i .

Eq 1 applies to isotopically homogeneous uranium where the relative efficiency is only dependent on energy.

3.3 The half-lives $T_{1/2}$ and the branching ratios BR are known, published nuclear data. The full energy peak counting intensity $C(E)$ is determined from the gamma-ray spectrum of the measured item.

3.4 The relative detection efficiency, $RE(E)$, is a function of gamma-ray energy and arises from the combined effects of detector response, attenuation due to absorbers and container walls, and self-absorption within the measured item for gamma rays of differing energies. The relative detection efficiencies are determined for each measured item from the observed gamma-ray spectrum by considering a series of gamma rays from a single isotope. The quotient of the photopeak counting intensity for gamma ray j with energy E_j emitted from isotope i and the branching ratio of gamma ray j from isotope i is proportional to the relative detection efficiency at energy E_j . This quotient defines the shape of the relative efficiency as a function of energy.

$$\frac{C(E_j^i)}{BR_j^i} \propto \left(\frac{N^i}{T_{1/2}^i} \right) \cdot RE(E_j) \quad (2)$$

⁴ Available from American National Standards Institute (ANSI), 25 W. 43rd St., 4th Floor, New York, NY 10036, <http://www.ansi.org>.

Relative efficiency data from different isotopes may be used to extend the relative efficiency curve. Data from different isotopes have the same shape but different normalization constants. It is customary to fit all the relative efficiency data to a mathematical function. The mathematical function is often chosen to represent the physical processes present in the measurement although simple polynomial functions have also been used. This process relies on the assumption that only a single isotopic composition is present.

3.5 All factors in Eq 1 are either determined from the gamma-ray spectrum of the measured item or are known, published nuclear constants. The isotope amount ratios are determined without recourse to standards or calibration by this so-called Intrinsic Calibration technique.

3.6 The measured isotope amount ratios can be converted to isotope amount fractions when the isotope amount ratios of all isotopes are measured with respect to a common isotope.

3.6.1 *Example*—Measuring isotope amount ratios (Eq 1) with respect to ²³⁸U. Measure f_{234}/f_{238} , f_{235}/f_{238} , ... f_i/f_{238} , where f_i is the isotope amount fraction of isotope i in the measured item.

3.6.2 All isotope amount fractions must sum to unity.

$$1.0 = f_{234} + f_{235} + f_{236} + f_{238} \quad (3)$$

3.6.3 Dividing Eq 3 by the common isotope amount fraction, f_{238} , and rearranging gives an expression for the isotope amount fraction of the common isotope, ²³⁸U here, in terms of the directly measured isotope amount ratios (see 3.6.1).

$$f_{238} = \left[\frac{f_{234}}{f_{238}} + \frac{f_{235}}{f_{238}} + \left(\frac{f_{236}}{f_{238}} \right) + 1 \right]^{-1} \quad (4)$$

3.6.4 Compute the other isotope amount fractions from the measured isotope amount ratios and Eq 4.

$$f_i = \frac{f_i}{f_{238}} \times f_{238}, i \neq 236 \quad (5)$$

3.6.5 Compute f_{236} from an isotopic correlation or acceptable knowledge. (See 1.4 and Refs (1 and 2).)

3.6.6 Renormalize, if needed, so all isotope amount fractions sum to unity.

3.6.7 The directly measured isotope amount fractions may be converted to isotope mass fractions, if desired, using:

$$m_{xxx} = \frac{f_{xxx} \cdot A_{xxx}}{(f_{234} \cdot A_{234} + f_{235} \cdot A_{235} + f_{236} \cdot A_{236} + f_{238} \cdot A_{238})} \quad (6)$$

where:

m_{xxx} = isotope mass fraction of isotope xxx,

A_{xxx} = atomic mass of isotope xxx, and

f_{xxx} = directly measured isotope amount fraction of isotope xxx.

4. Significance and Use

4.1 The determination of uranium isotopic composition by gamma-ray spectrometry is a nondestructive technique and when used with other nondestructive techniques that quantify a single isotope, such as Test Methods C1133 (Segmented Gamma Scanning), C1221 (Solution Assay), and C1451 (Point Assay),

and C1718 (Tomographic Gamma Scanning), can provide a wholly nondestructive assay of uranium mass necessary for material accountancy and safeguards needs. This method can be used with calorimetry (Test Method C1458) for kilogram quantities of high-enriched uranium and is also used to convert an Active-Well Coincidence Counter (4) measurement of ^{235}U mass to total uranium mass.

4.2 Because gamma-ray spectrometry systems are typically automated, the routine use of the test method is fast, reliable, and is not labor intensive. The test method is nondestructive, requires no sample preparation, and does not create waste disposal problems.

4.3 The test method does not require that the system be calibrated to a specific geometry.

4.4 The test method assumes that all uranium in the measured item has the same isotopic distribution. This is often termed isotopic homogeneity.

4.5 The application of the test method does not depend upon the physical or chemical form of the material being analyzed.

4.6 The ^{236}U abundance is not measured by this test method and must be estimated from isotopic correlation techniques, stream averages, historical information, or other measurement techniques.

4.7 The isotopic composition of a given item of uranium is an attribute of that item and, once determined, can be used in subsequent inventory measurements to verify the identity of an item within the measurement uncertainties.

4.8 The method can also measure the ratio of other gamma-emitting isotopes in the measured item to uranium assuming they have the same spatial distribution as the uranium in the item. Some of these “other” gamma-emitting isotopes include daughter isotopes of uranium, cesium, and other fission products.

4.9 The method can be applied to gamma and x rays in two overlapping energy regions, depending upon the nature of the measured item, its containment, and the characteristics of the detector used for data acquisition.

4.9.1 60 keV to 250 keV—This energy range requires good energy resolution provided by planar or semi-planar HPGe detectors. The analysis methods must be capable of deconvoluting the x-ray peak line shapes from the gamma-ray peak shapes.

4.9.2 120 keV to 1010 keV—This energy range generally requires higher efficiency detectors typified by larger coaxial detectors (> 25 % relative efficiency) or large semi-planar detectors (> 30 mm thick).

4.10 Fig. 1 shows the decays that produce most of the prominent gamma and x rays that are measured in this analysis.

4.11 The energies and intensities of the principal gamma rays and x rays used in isotopic analysis of uranium by gamma spectrometry are shown in Table 1. The data in Table 1 are indicative. Individual codes may use slightly different numbers empirically adjusted to give good performance in conjunction with the overall implementation of the code.

5. Interferences

5.1 Because of the finite resolution of even the best quality HPGe detectors, the presence of other gamma-emitting sources must be assessed for their effects on the isotopic abundance determination.

5.1.1 The detector used for the spectral measurements shall be adequately shielded from other nearby radiation sources. Background spectra should be collected to ensure the effectiveness of detector shielding and to identify the background radiations.

5.1.2 If fission products are present in the item being measured, they will contribute additional gamma-ray spectral peaks. These peaks occur mainly in the 500 keV to 800 keV energy range and may affect the intensity determination of uranium-related peaks in this region. These high-energy gamma rays from fission products also produce contributions to the Compton background below 500 keV that decrease the precision for peak intensity determination in this region.

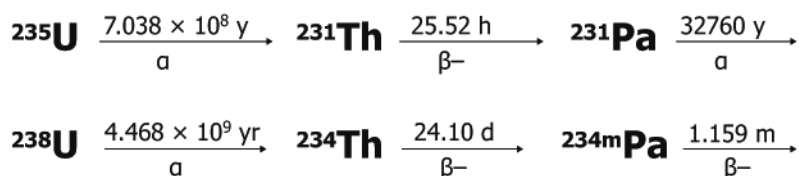
5.1.3 Plutonium gamma rays commonly interfere with uranium gamma rays in mixed uranium-plutonium oxides (MOX). The analysis of MOX is discussed in Test Method C1030.

5.1.4 Trace levels of Th daughter products are often found in uranium spectra from uranium materials that have a reactor history.

5.1.4.1 These Th daughter products arise from the decay of reactor-produced ^{232}U that may be present in mass fractions of ^{232}U to total U of 10^{-10} g/g or less. Th daughter gamma rays are most prominent for uranium with ^{235}U enrichment > 10 % and their intensity generally increases as enrichment increases.

5.1.4.2 Uranium enriched directly from natural uranium that has never been in a reactor will not contain Th daughter gamma-ray peaks.

5.1.4.3 Th daughter product gamma rays may be used to provide relative efficiency information (Eq 2) in energy regions (200 keV to 900 keV) where there are very few uranium gamma rays. Useful Th daughter gamma rays are listed in Table 2. Weaker Th daughter gamma rays (727 keV, 763 keV) may interfere with neighboring ^{238}U daughter gamma rays and must be accounted for in analyses in that energy region.



NOTE 1—Nuclear Data from Ref (1).

FIG. 1 Decay of ^{235}U and ^{238}U

TABLE 1 Energies and Branching Intensities of Principal X- and Gamma-Ray Peaks Used in Gamma-Ray Isotopic Analysis of Uranium^A

Isotope	Energy (keV)	Branching Intensity (photons/disintegration, %)
²³¹ Th	81.2280 (14) ^B	0.90 (5)
²³¹ Th	84.2140 (13)	6.6 (4)
ThK α 2	89.957	3.43 (9)
PaK α 2	92.282	0.35 (4)
²³⁴ Th	92.38 (1)	2.13 (20)
²³⁴ Th	92.80 (2)	2.10 (20)
ThK α 1	93.350	5.54 (14)
UK α 2	94.654	0.626 ^C
PaK α 1	95.863	0.57 (6)
UK α 1	98.434	1.000 ^C
²³⁴ U	120.90 (2)	0.035 (5)
²³⁵ U	143.76 (2)	10.96 (14)
²³⁵ U	163.356 (3)	5.08 (6)
²³⁵ U	185.715 (5)	57.0 (6)
²³⁵ U	202.12 (1)	1.080 (23)
²³⁵ U	205.316 (10)	5.02 (6)
^{234m} Pa	258.227 (3)	0.0764 (21)
^{234m} Pa	742.813 (5)	0.1066 (23)
^{234m} Pa	766.42 (10)	0.317 (5)
^{234m} Pa	786.28 (10)	0.0544 (8)
^{234m} Pa	1001.03 (10)	0.842 (8)

^A Energies and Branching Intensities from Ref (1).

^B Uncertainties in parentheses are absolute 1 σ values.

^C Relative values from unweighted mean of plutonium decay data from Ref (1).

TABLE 2 Useful ²²⁸Th Daughter Gamma Rays^A

Energy (keV)	Source	Branching Fraction
238.632 (2)	²¹² Pb	0.436 (5)
240.986 (6)	²²⁴ Ra	0.0410 (5)
277.371 (5)	²⁰⁸ Tl	0.0237 (11)
300.087 (10)	²¹² Pb	0.0330 (4)
583.187 (2)	²⁰⁸ Tl	0.3055 (11)
722.04 (12)	²⁰⁸ Tl	8.6 (14) E-04
727.330 (9)	²¹² Bi β -	0.0667 (9)
763.13 (8)	²⁰⁸ Tl	6.43 (11) E-03
785.37 (8)	²¹² Bi β -	0.01102 (13)
860.557 (4)	²⁰⁸ Tl	0.04493 (36)

^A Branching Fraction and Energy from Ref (1).

5.1.5 Samples with Th mixed with U would contribute additional x-ray fluorescence from Th which must be accounted for in analyses in the 60 keV to 250 keV region.

5.1.6 Uranium ore samples produce many gamma rays and x rays from ²¹⁴Pb, ²¹⁴Bi, and other decay products that can interfere with the uranium analysis peaks.

5.1.7 Bremsstrahlung from the β^- decay of ^{234m}Pa contributes greatly to the background continuum for items with high concentrations of ²³⁸U. This adversely affects the signal-to-background ratio, especially for gamma ray peaks below 500 keV.

5.2 Count rate and coincidence summing effects may also affect the isotopic abundance determination.

5.2.1 True coincidence summing effects increase as sample-to-detector distance decreases and also increase as the detector diameter increases.

5.2.2 True coincidence summing affects measurements in the 120 keV to 1010 keV range for measured-item to detector distances $< \sim 12$ cm using a ~ 55 mm diameter coaxial detector. These effects can bias the ²³⁵U/²³⁸U amount ratio by as much as 15 % (5).

5.2.2.1 These effects can be corrected (5) and current generation analysis software (2) applies these corrections in a transparent fashion.

5.2.3 True coincidence summing effects are also observed for analyses in the 60 keV to 250 keV region for an analysis code using the ThK α 1 and ThK α 2 x-ray peaks for ²³⁵U analysis (6). These effects bias the results by 5 % to 8 % for detector-sample contact and essentially vanish at sample-detector distance of 10 cm.

5.2.3.1 Analysis software that minimizes the use of Th x rays for ²³⁵U analysis does not exhibit this effect for analysis in the 60 keV to 250 keV region.

5.2.4 Random summing and pileup can affect measurements in the 120 keV to 1010 keV energy range and also the 60 keV to 250 keV energy range.

5.2.4.1 The 163.36 keV ²³⁵U gamma ray can sum with UK α x rays to produce interferences around the 258.3 keV ²³⁸U daughter peak which is important in 120 keV to 1010 keV energy-range analyses. The software of Ref (2) corrects for this effect.

5.2.4.2 The 143.76 keV ²³⁵U gamma ray can sum with UK α x rays to produce interferences around the Th daughter gamma ray at 238.6 keV which can be used for relative efficiency determination in 120 keV to 1010 keV energy range analysis. The software of Ref (2) corrects for this effect.

5.2.4.3 Lead K α x rays (72.8 keV, 75.0 keV) produced in Pb shielding around the detector, can sum with the strong ²³⁵U gamma ray at 185.72 keV to produce interferences in the 258 keV region. This effect can be removed by properly shielding the detector from Pb x rays with a graded-Z filter that lines the inside of the Pb detector shield.

5.2.4.4 Lead K β x rays around 84.8 keV and 87.5 keV can interfere with the 84.22 keV ²³¹Th peak possibly affecting analysis in the 60 keV to 250 keV energy range. This effect can be removed by properly shielding the detector from Pb x-rays (typically from Pb shielding around the detector) with a graded-Z filter lining the Pb shielding.

6. Apparatus

6.1 *Cooled High-Purity Germanium Detector, Preamplifier*—Cooling of the HPGe crystal may come from liquid nitrogen (LN₂) or from electric or electro-mechanical coolers that do not use LN₂. The configuration of the HPGe detector may be planar, semi-planar, or coaxial with the type, size and energy resolution of the detector chosen to accommodate the energy range of analysis for the desired measurements. Planar or semiplanar detectors with energy resolution (full-width at half-maximum) at 122 keV better than 700 eV are best for analysis of spectra in the 60 keV to 250 keV region. Larger volume coaxial detectors with relative efficiencies > 25 % (ANSI/IEEE Std 325-1996) or large volume semi-planar detectors (4.9.2) are used for analysis in the 120 keV to 1010 keV energy region. Resolution of 2 keV or better at 1332 keV is preferred.

6.2 *High Voltage Supply, Linear Amplifier, Analog-to-Digital Converter (ADC), Multichannel Pulse-Height Analyzer (MCA)*—Systems may consist of these individual components, or a preferred and more convenient commercial integrated