Designation: C1030 – 03

Standard Test Method for Determination of Plutonium Isotopic Composition by Gamma-Ray Spectrometry[1]

This standard is issued under the fixed designation C1030; 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 is applicable to the determination of isotopic abundances in isotopically homogeneous Pu-bearing materials. This test method may be applicable to other plutonium-bearing materials, some of which may require modifications to the described test method.

1.2 The procedure is applicable to sample sizes ranging from a few tenths of a gram up to the maximum sample weight allowed by criticality limits.

1.3 Because ^{242}Pu has no useful gamma-ray signature, its isotopic abundance is not determined. Isotopic correlation techniques may be used to estimate its relative abundance (Refs **1, 2**).[2]

1.4 This test method has been demonstrated in routine use for isotopic abundances ranging from 96 to 55 % ^{239}Pu. This test method has also been employed for isotopic abundances outside this range.

1.5 The values stated in SI units are to be regarded as the standard.

1.6 *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:*[3]

C697 Test Methods for Chemical, Mass Spectrometric, and Spectrochemical Analysis of Nuclear-Grade Plutonium Dioxide Powders and Pellets

C698 Test Methods for Chemical, Mass Spectrometric, and Spectrochemical Analysis of Nuclear-Grade Mixed Oxides ((U, Pu)O_2)

C982 Guide for Selecting Components for Energy-Dispersive X-Ray Fluorescence (XRF) Systems[4]

C1207 Test Method for Nondestructive Assay of Plutonium in Scrap and Waste by Passive Neutron Coincidence Counting

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

C1500 Test Method for Nondestructive Assay of Plutonium by Passive Neutron Multiplicity Counting

E181 Test Methods for Detector Calibration and Analysis of Radionuclides

E267 Test Method for Uranium and Plutonium Concentrations and Isotopic Abundances

2.2 *ANSI Standards:*

ANSI N15.35 Guide to Preparing Calibration Material for Nondestructive Assay Systems that Count Passive Gamma Rays[5]

3. Summary of Test Method

3.1 Relative intensities of gamma-rays from a plutonium sample are determined from a gamma-ray spectrum obtained with a high-resolution Ge detector.

3.2 The atom ratio, N_i/N_j, for isotopes i and j is related to the relative counting intensities, I_i and I_j, for the gamma-rays of energy E_i and E_j by:

$$\frac{N_i}{N_j} = C_{ij}\frac{I_i}{\varepsilon_i}\cdot\frac{\varepsilon_j}{I_j} \quad (1)$$

$$C_{ij} = \frac{T^{1/2}_i}{T^{1/2}_j}\cdot\frac{B_j}{B_i} \quad (2)$$

[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.

[4] Withdrawn. The last approved version of this historical standard is referenced on www.astm.org.

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

where:

ε = relative detection efficiency for a gamma-ray at energy E,
$T^{1/2}$ = half-life, and
B = gamma-ray branching intensity (usually expressed as the gamma-ray emission probability per disintegration).

3.3 The conversion factors, C_{ij}, are computed from known half-lives and gamma-ray branching intensities.

3.4 The relative detection efficiency, ε, is a function of gamma-ray energy and results from the combined effects of detector response, attenuation due to absorbers and container walls, and self-absorption within the sample for gamma-rays of differing energies. The relative detection efficiencies are determined for each sample from the observed gamma spectrum.

4. Significance and Use

4.1 The determination of isotopic composition by gamma-ray spectrometry is a nondestructive technique and when used with other nondestructive techniques, such as calorimetry (Test Method C1458) or neutron counting (Test Methods C1207, C1493, and C1500), can provide a totally nondestructive plutonium assay necessary for material accountancy and safeguards needs.

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

4.3 This test method assumes that the isotopic composition of plutonium in the sample being measured is homogeneous (see see 7.2.4 and (5)).

4.4 The ^{242}Pu abundance is not measured by this test method and must be estimated from isotopic correlation techniques, stream averages, historical information, or other measurement techniques.

4.5 A daughter product of ^{241}Pu is ^{241}Am. The ^{241}Am/^{239}Pu atom ratio can also be determined by means of this test method (assuming a homogeneous isotopic distribution of plutonium and ^{241}Am) and is necessary for the correct interpretation of a calorimetric heat measurement.

4.6 The isotopic composition of a given batch or sample of plutonium is an attribute of that sample and, once determined, can be used in subsequent inventory measurements to verify the identity of a sample within the measurement uncertainties.

4.7 The method can also measure the ratio of other gamma emiting isotopes to plutonium assuming they have the same spatial distribution as the plutonium in the sample. Some of these "other" gamma-emitting isotopes include isotopes of uranium, neptium, curium, cesium, and other fission products. (The same methods of this standard can be used to measure the isotopic composition of uranium in samples containing only uranium (**4–6**)).

5. Interferences

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

5.1.1 The germanium detector used for the spectral measurements shall be adequately shielded from other nearby plutonium sources. Background spectra shall 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 sample being measured, they will contribute additional gamma-ray spectral peaks. These peaks occur mainly in the 500 to 800-keV energy range and may affect the intensity determination of plutonium and americium 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 For mixed plutonium-uranium oxide samples, the appropriate corrections for the spectral peaks produced by uranium gamma emission shall be applied. The main interferences due to uranium are listed in Table 1.

5.1.4 Other interference-producing nuclides can be routinely present in plutonium-bearing materials. The gamma rays from these nuclides must be assessed for their interference effects on the multiplets used for the plutonium isotopic analysis and the proper spectral corrections applied. Some of these interfering nuclides would include: ^{237}Np and its daughter ^{233}Pa, ^{239}Np, ^{243}Am, and ^{233}U.

5.2 Count-rate and coincident summing effects may also affect the isotopic abundance determination. This is especially important for samples having high americium concentrations (typically greater than ^{241}Pu ingrowth). Summing of the intense 59.5-keV transition with other intense gamma radiations produces spurious spectral peaks (**7**). Thin (typically 0.5 to 2 mm) cadmium or tin (which is less toxic) absorbers shall be placed on the front face of the detector to keep the height of the 59.5 keV gamma-ray peak equal to or less than the height of the most intense peaks in the 100-keV region.

6. Apparatus

6.1 *Germanium Detector (with liquid nitrogen supply), Preamplifier and High-Voltage Supply*—Energy resolution of the detector for spectra collected below 400 keV should be better than 600 eV full-width-at-half-maximum (FWHM) at 122 keV. Purchase specifications of 550 eV or less should ensure a working resolution of 600 eV or better. These detectors are generally intrinsic, planar Ge of a few cubic centimeters active volume. For the energy regions above 400 keV, a large volume Ge detector with an active volume of 40 cm^3 or greater and with resolution of 2.0 keV or better at 1332 keV is preferred.

TABLE 1 Gamma-Ray Interferences Due to Uranium in (Pu, U)O_2 Materials

Energy (keV)	Branching Intensity (%γ/disintegration)	Isotope
143.77	10.7	^{235}U
163.36	4.85	^{235}U
185.72	56.1	^{235}U
202.12	1.07	^{235}U
205.31	4.87	^{235}U

6.2 *Linear Amplifier, Analog-to-Digital Converter (ADC), Multichannel Pulse-Height Analyzer (MCA)*—The ADC-MCA combination shall be capable of at least 4K channel conversion and storage. More detailed descriptions of these components can be found in Guide C982.

6.3 High count rate applications require the use of pile-up rejection circuitry. Digital stabilization may be desirable for long count times or poor environmental control to ensure the quality of the spectral data.

6.4 Because of the complexity of plutonium spectra, data reduction is usually performed by computer. Several software codes are available that perform the spectral analysis and isotopic abundance calculations on a computer (Refs **8–11**).

6.5 All of the above apparatus is commercially available. Electronic modules are either NIM standard, NIM compatible, or self-contained, fully integrated digital signal processing units. Many gamma-ray spectrometry systems are interfaced to a computer. This permits the isotopic abundance determination procedure to be automated.

7. Precautions

7.1 *Safety Precautions*—Plutonium-bearing materials are both radioactive and toxic. Use adequate laboratory facilities and safe operating procedures in handling samples containing these materials. Safe handling practices are outlined in References (**12-14**).

7.2 *Technical Precautions*:

7.2.1 Preclude or rectify counting conditions that may produce spectral distortions. Use pulse pile-up rejection techniques if high count rates are encountered. Use absorbers when appropriate to reduce the intensity of the 59.5 keV gamma-ray of americium (see 5.2). Temperature and humidity fluctuations in the measurement environment may cause gain and zero-level shifts in the gamma-ray spectrum. Employ environmental controls or digital stabilization, or both, in this case. Failure to isolate the electronic components from other electrical equipment or the presence of noise in the AC power may also produce spectral distortions.

7.2.2 The alpha decay branch of ^{241}Pu proceeds through the daughter ^{237}U, which in turn decays to ^{237}Np with a half-life of 6.75 days. About eight weeks are required for secular equilibrium to be achieved. If less than eight weeks have elapsed since separation, use gamma rays produced by the parent, ^{241}Pu, for isotopic abundance determinations; for example, the 148.57 keV peak. However, gamma rays arising from decay of the daughter, ^{237}U, can be used for relative efficiency calculations.

7.2.3 Preferably, do not include high-Z absorbers in sample packaging. As little as ⅛ in. (0.32 cm) of lead surrounding the plutonium will absorb the majority of the useful gamma rays in the 100 to 200-keV region and may invalidate the measurement.

7.2.4 The isotopic composition of all the plutonium in the sample must be the same. The technique does not apply to nonuniform mixtures of different isotopic composition. However, the physical distribution of the plutonium within the sample may be nonuniform with no adverse effect on the results.

7.2.5 The ^{241}Am/^{239}Pu atom ratio must be uniform in all the plutonium in the sample, in order to obtain reliable specific power measurements to use in interpreting calorimetry results. Certain types of Pu materials with nonhomogeneous Am-Pu distributions (salt residues) have been shown to be amenable to assay by this test method with slight modifications **(15, 16).** These materials have a low density salt matrix containing most of the americium while most of the plutonium is dispersed throughout this matrix as high density localizations or free metal shot.

7.2.6 Plutonium-bearing materials, especially plutonium fluoride compounds, should not be stored in the vicinity of, or on, the germanium detectors. High energy neutrons emitted by these materials can produce trapping centers in the germanium crystals and severely degrade the resolution of the detectors. The use on N-type detectors which are less susceptible to neutron damage, can prolong useful detector life.

8. Calibration, Standardization, and Measurement Control

8.1 *Apparatus*—The energy calibration of the spectrometry system can be adjusted using a gamma-emitting check source or a plutonium-bearing sample because the plutonium gamma-ray energies are well known. A listing of the intense plutonium radiations that are suitable for an energy calibration procedure is given in Table 2. See also Test Methods E181 and Reference **17.**

8.2 *Reference Materials*:

8.2.1 The expression relating atom ratios to detected peak intensities contains only fundamental constants (see Eq 1 and Eq 2) and does not depend upon reference standards. Reference standards can be used to identify biases in the values of measured fundamental constants and as an aid in identifying possible spectral interferences.

TABLE 2 Energies and Gamma-Ray Branching Intensities[A] of Prominent Pu and Am Spectral Peaks

Energy (keV)	Branching Intensity (γ/disintegration, %)	Isotope
59.54	0.359	^{241}Am
125.29	4.08×10^{-5}	^{241}Am
129.29	6.26×10^{-5}	^{239}Pu
148.57	1.87×10^{-6}	^{241}Pu
152.68	9.37×10^{-6}[B]	^{238}Pu
160.28	4.02×10^{-6}	^{240}Pu
164.48[C]	4.53×10^{-7}	^{241}Pu-^{237}U
	6.67×10^{-7}	^{241}Am
203.54	5.60×10^{-6}	^{239}Pu
208.00[C]	5.16×10^{-6}[B]	^{241}Pu-^{237}U
	7.91×10^{-6}	^{241}Am
335.44[C]	2.39×10^{-8}	^{241}Pu-^{237}U
	4.96×10^{-6}	^{241}Am
345.01	5.59×10^{-6}	^{239}Pu
368.61[C]	1.05×10^{-8}	^{241}Pu-^{237}U
	2.17×10^{-6}	^{241}Am
375.04	1.57×10^{-5}	^{239}Pu
413.71	1.49×10^{-5}	^{239}Pu
642.48	1.245×10^{-7}	^{240}Pu
645.97	1.49×10^{-7}	^{239}Pu
662.42	3.64×10^{-6}	^{241}Am
717.72	2.74×10^{-8}	^{239}Pu
721.99	1.96×10^{-6}	^{241}Am

[A] Branching intensities from Ref **15**, except where noted.
[B] Branching intensity from "Handbook of Nuclear Data for Safeguards," INDC (NDS)-248, Nuclear Data Section, IAEA, Vienna, Austria, 1991.
[C] Produced in decay of ^{241}Pu-^{237}U and ^{241}Am, total intensity will be a function of the abundances of these two isotopes.