Designation: C1030 – 10

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 plutonium-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 items containing plutonium masses ranging from a few tens of milligrams up to the maximum plutonium mass allowed by criticality limits.

1.3 Measurable gamma ray emissions from plutonium cover the energy range from approximately 30 keV to above 800 keV. K-X-ray emissions from plutonium and its daughters are found in the region around 100 keV. This test method has been applied to all portions of this broad spectrum of emissions.

1.4 The isotopic abundance of the ^{242}Pu isotope is not directly determined because it has no useful gamma-ray signature. Isotopic correlation techniques may be used to estimate its relative abundance Refs (**1**) and (**2**).[2]

1.5 This test method has been demonstrated in routine use for isotopic abundances ranging from 99 to <50 % ^{239}Pu. This test method has also been employed for isotopic abundances outside this range.

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

1.7 *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 (Withdrawn 2008)[4]

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

C1316 Test Method for Nondestructive Assay of Nuclear Material in Scrap and Waste by Passive-Active Neutron Counting Using ^{252}Cf Shuffler

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:*[5]

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

[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] 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.

3. Summary of Test Method

3.1 The intensities of gamma-rays emitted from a plutonium-bearing item are determined from a gamma-ray spectrum obtained with a High-Purity Germanium (HPGe) detector. The method has also been used with CdTe detectors.

3.2 The atom ratio, N^i/N^k, for isotopes i and k is related to the photopeak counting intensity, $C\ (E_j^i)$, for gamma ray j with energy E_j emitted from isotope i by:

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

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.

3.3 The half lives $T_{1/2}$ and the branching ratios BR are known, published nuclear data. The photopeak 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 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.

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

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 absolute atom ratios are determined without recourse to standards or calibration by this so-called Intrinsic Calibration technique.

4. Significance and Use

4.1 The determination of plutonium 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, C1316, C1493, and C1500), can provide a wholly nondestructive plutonium assay necessary for material accountancy and safeguards needs.

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 This test method assumes that all plutonium in the measured item has the same isotopic distribution, often called isotopic homogeneity (see 7.2.4 and 7.2.5).

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 Americium-241 is a daughter product of ^{241}Pu. 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). The determination of the ^{241}Am/^{239}Pu atom ratio is necessary for the correct interpretation of a calorimetric heat measurement.

4.6 The isotopic composition of a given batch or item of plutonium 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.

4.7 The method can also measure the ratio of other gamma-emitting isotopes to plutonium assuming they have the same spatial distribution as the plutonium in the item. Some of these "other" gamma-emitting isotopes include isotopes of uranium, neptunium, curium, cesium, and other fission products. The same methods of this standard can be used to measure the isotopic composition of uranium in items containing only uranium **(3, 4, 5, 6)**.

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 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 item 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-bearing items, the appropriate corrections for the spectral peaks produced by uranium gamma emission shall be applied. The main interferences from 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 include: ^{237}Np and its daughter ^{233}Pa, ^{243}Am and its daughter ^{239}Np, ^{233}U, and the Th decay chain daughters of ^{232}U and ^{236}Pu.

TABLE 1 Principal Gamma-Ray Interferences from Uranium in Mixed Pu/U Materials[A]

Energy (keV)	Branching Intensity γ/disintegration, (%)	Isotope
143.76	10.96	^{235}U
163.33	5.08	^{235}U
185.715	57.2	^{235}U
202.11	1.08	^{235}U
205.311	5.01	^{235}U

[A] Branching Intensity and Energy from Ref (7).

5.2 Count-rate and coincident-summing effects may also affect the isotopic abundance determination. This is especially important for items having high ^{241}Am concentrations. Random summing of the intense 59.5-keV ^{241}Am gamma ray with other intense gamma radiations produces spurious spectral peaks (8) that can interfere with the isotopic analysis. Thin (typically 0.5 to 2 mm) cadmium or tin (which is less toxic) absorbers should 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 *Cooled High-Purity Germanium Detector, Preamplifier*—Cooling of the HPGe crystal may come from liquid nitrogen (LN_2) or from electric or electro-mechanical coolers that do not use LN_2. 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 semi-planar detectors with energy resolution (full-width at half maximum) at 122 keV better than 650 eV are best for analysis of spectra in the 60 to 450 keV region. Larger volume coaxial detectors with efficiencies (relative to a 3 × 3 NaI(Tl) at 1332 keV for a point source at a distance of 10 cm (ANSI/IEEE Std 325-1996)) of 25 to 100 % are used for analysis in the energy regions above 120 keV. 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 containing these components compliant with Guide C982 may be used. A preferred and more convenient choice is an integrated digital spectroscopy system containing all components in a single unit with a high speed computer interface. Analysis of spectra in the 100 keV-region requires at least 4096 channels of data. Analysis in higher energy regions requires a minimum of 8192 channels of data with 16 384 data channels becoming more widely used.

6.3 High count rate applications require the use of pile-up rejection circuitry. Digital stabilization may be desirable for long count times under conditions of poor environmental control to ensure the quality of the spectral data. High quality digital spectroscopy systems fulfill all of these requirements and have been shown to have minimal degradation on plutonium isotopic composition measurement results at input counting rates as high as 100 kHz (9).

6.4 Because of the complexity of plutonium spectra, data reduction is usually performed by computer. Computerized analysis methods are well developed and have been highly automated with the development of various analysis software codes (9, 10, 11, 12, 13, 14, 15). Analysis software is commercially available as are all of the required data acquisition components.

7. Precautions

7.1 *Safety Precautions*—Plutonium-bearing materials are both radioactive and toxic. Use adequate laboratory facilities and safe operating procedures in handling items containing these materials. Follow all safe operating procedures and protocols specific to the facility or location where the measurements are being made.

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 decay of ^{241}Pu is shown in Fig. 1. The alpha decay branch proceeds through the daughter ^{237}U which decays with a 6.75 day half-life to ^{237}Np. It takes 67 days to reach 99.9 % of secular equilibrium for this branch of the decay. After secular equilibrium has been attained the strong gamma rays at 164.6, 208.0, 267.5, 332.4, 335.4, 368.6, and 370.9 keV from the decay of ^{237} U may be used to directly determine ^{241}Pu. These major gamma rays from the decay of ^{237}U also have an identical energy component from the beta decay branch of ^{241}Pu proceeding through ^{241}Am. The ^{241}Am component of these "co-energetic" peaks must be accounted for in the analysis. If secular equilibrium has not been reached, gamma rays (usually the 148.57-keV peak) from the direct decay of ^{241}Pu to ^{237}U must be used to determine the ^{241}Pu isotopic fraction. At all times the gamma rays from the decay of ^{237}U may be used to determine the relative efficiency.

7.2.3 The facility may place high-Z absorbers within the sealed, plutonium-bearing container to reduce external radiation exposure to the handler. As little as 1⁄16 in. (0.16 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, depending upon the energy range of the analysis.

7.2.4 The isotopic composition of all the plutonium in the item must be the same. The technique does not apply to nonuniform or heterogeneous mixtures of different isotopic composition. However, the physical distribution or chemical composition of the plutonium within the item 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 item, in order to obtain reliable specific power