Designation: C 1030 – 03

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

This standard is issued under the fixed designation C 1030; 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:*

C 697 Test Methods for Chemical, Mass Spectrometric, and Spectrochemical Analysis of Nuclear-Grade Plutonium Dioxide Powders and Pellets[3]

C 698 Test Methods for Chemical, Mass Spectrometric, and Spectrochemical Analysis of Nuclear-Grade Mixed Oxides ((U, Pu)O_2)[3]

C 982 Guide for Selecting Components for Energy-Dispersive X-Ray Fluorescence (XRF) Systems[3]

C 1207 Test Method for Nondestructive Assay of Plutonium in Scrap and Waste by Passive Neutron Coincidence Counting[3]

C 1458 Test Method for Nondestructive Assay of Plutonium, Tritium and^{241}Am by Calorimetric Assay[3]

C 1493 Test Method for Non-Destructive Assay of Nuclear Material in Wast by Passive and Active Neutron Counting Using a Differential Die-Away System[3]

C 1500 GTest Method for Nondestructive Assay of Plutonium by Passive Neutron Multiplicity Counting[3]

E 181 General Methods for Detector Calibration and Analysis of Radionuclides[4]

E 267 Test Method for Uranium and Plutonium Concentrations and Isotopic Abundances[4]

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:

$$\frac{N_i}{N_j} = C_{ij}\frac{I_i}{\epsilon_i}\cdot\frac{\epsilon_j}{I_j} \tag{1}$$

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

where:

ϵ = relative detection efficiency for a gamma-ray at energy E,

---

[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 Nondestructive Assay.

Current edition approved July 10, 2003. Published August 2003. Originally approved in 1984. Last previous edition approved in 2001 as C 1030 – 95(2001).

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

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

[4] *Annual Book of ASTM Standards*, Vol 12.02.

[5] Available from the American National Standards Institute, 11 W. 42nd St., 13th Floor, New York, NY 10036.

$T^{1/2}$ = 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 C 1458) or neutron counting (Test Methods C 1207C 1493C 1500), 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.

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

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

and storage. More detailed descriptions of these components can be found in Guide C 982.

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 E 181 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.

8.2.2 Working reference materials with isotopic composition traceable to the National Institute of Standards and Technology (NIST) (or other certifying standards bodies)

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.

reference materials may be used to verify the overall correct operation of the spectrometry system and data reduction techniques, and also as an aid in identifying interferences. Currently available NIST isotopic reference materials (SRMs NIST-946, NIST-947, and NIST-948) are usually not suitable for direct use as reference standards in all cases due to the small plutonium mass of the materials. Working reference materials traceable to the NIST standard reference materials should be prepared and validated by other analysis techniques (see Test Methods C 697, C 698, and E 267).

8.3 *Measurement Control*:

8.3.1 A measurement control program shall be established in order to identify anomalous measurement results that may be due to instrument failure or operator (procedural) error. A pragmatic measurement control program will involve tradeoffs between operator convenience, cost, throughput, and the ability of the program to detect all possible failures. The factors that may affect the degree of automation of the measurement control program are: the expertise level of the operator, the customer requirements, and the type of isotopic measurement being made. The measurement control program shall cover all phases of the plutonium isotopic measurement from the data collection through the calculation of the isotopic atom ratios.

8.3.2 Data collection procedures shall be standardized for each type of sample or measurement application. Control limits or ranges shall be established for the various data collection parameters such as: count time, count rate, system dead time, and counting geometry. To assure the quality of the collected data and analysis methods, the isotopic measurement control program would employ both the internal and external techniques or checks discussed below.

8.3.3 Internal checks utilize parameters or measurement results from the spectral data of the sample item being assayed. An important internal check that provides a good indication of the overall hardware performance is the system resolution. The resolution of a certain peak, for example the 129.26 keV gamma ray of ^{239}Pu, should be monitored on a spectrum-to-spectrum basis. Depending upon the level of sophistication of the measurement control program, these values could be plotted on a control chart or compared against performance limits by the analysis software. In the latter case, if the system is determined to be out of control, the operator could be prevented by the software from continuing until remedial action is taken. Other internal checks that may be used are: the position of certain spectral peaks, and the precision of the plutonium atom ratios. Again, these parameters could be manually plotted on a control chart or could be monitored automatically by the measurement analysis software. If more than one peak pair is used to obtain a particular atom ratio, the internal consistency of the atom ratios, as determined by the various peak pairs, can be used as an indication of interferences that may affect measurement control. For analysis methods that use fitting techniques, the statistical measures of goodness-of-fit, such as chi-square, can be used with suitable control limits for measurement control purposes.

8.3.4 External checks rely on a comparison of isotopic results among replicate gamma-ray spectral measurements or between the spectral measurement and another assay technique. The isotopic assay of standard reference materials and working standards can be used to verify that the measurement system is still in control. Measurements of the same sample on parallel instruments can also be used as a measurement control indicator. Other external techniques are: comparisons of the gamma-ray results from a sample to destructive analysis results, participation in interlaboratory exchange programs, comparisons of the present data with historical or stream average data, and the reanalysis of samples at random.

8.3.5 A successful measurement control program will employ a combination of internal and external techniques. Total reliance on an individual technique or check is not recommended. The simpler measurement checks, such as the monitoring of the system resolution, should probably be performed on a sample-by-sample or daily basis, while other more complex techniques could be performed less frequently.

8.3.6 The measurement control data provided by the internal and external checks can be used for constructing a data base for identifying and monitoring the random and systematic errors associated with the isotopic measurement system.

TABLE 3 Pu and Am Half-Lives[A]

Nuclide	Half-Life (Years)
^{238}Pu	87.74 ± 0.04
^{239}Pu	24119 ± 26
^{240}Pu	6564 ± 11
^{241}Pu	14.348 ± 0.022
^{242}Pu	376300 ± 900
^{241}Am	433.6 ± 1.4

[A] Half-lives from Test Method C 1458..

9. Procedure

9.1 Arrange the counting geometry to obtain the maximum count rate that does not produce any unwanted spectral distortions. The 59.5 keV peak from ^{241}Am usually produces a substantial contribution to the system dead time; its intensity can be reduced through the use of a Cd or Sn absorber.

9.2 Acquire the spectrum for the length of time necessary to achieve the desired level of precision. The precision for an isotopic composition measurement depends on counting statistics and is a function of several parameters (see Section 11). Typical counting times are 10 min to 1h.

9.3 Determine the peak intensities from the pulse-height data. In principle, this involves subtracting the background continuum beneath the peak and removing any contributions from interfering adjacent peaks. Since plutonium spectra contain many complex multiplets, this data reduction is usually performed with the aid of a computer or microprocessor.

9.4 The uncertainties ascribed to the peak intensities shall be propagated from the statistical uncertainties of the measured peak areas and any uncertainties due to the peak area determination process. Specific examples would be method dependent (see Reference **18**).

10. Calculation

10.1 The isotopic abundance ratio for two isotopes is calculated using Eq 1. The use of an existing computer program to analyze the pulse-height data and obtain the isotopic abundances will obviate the need for making further calculations.

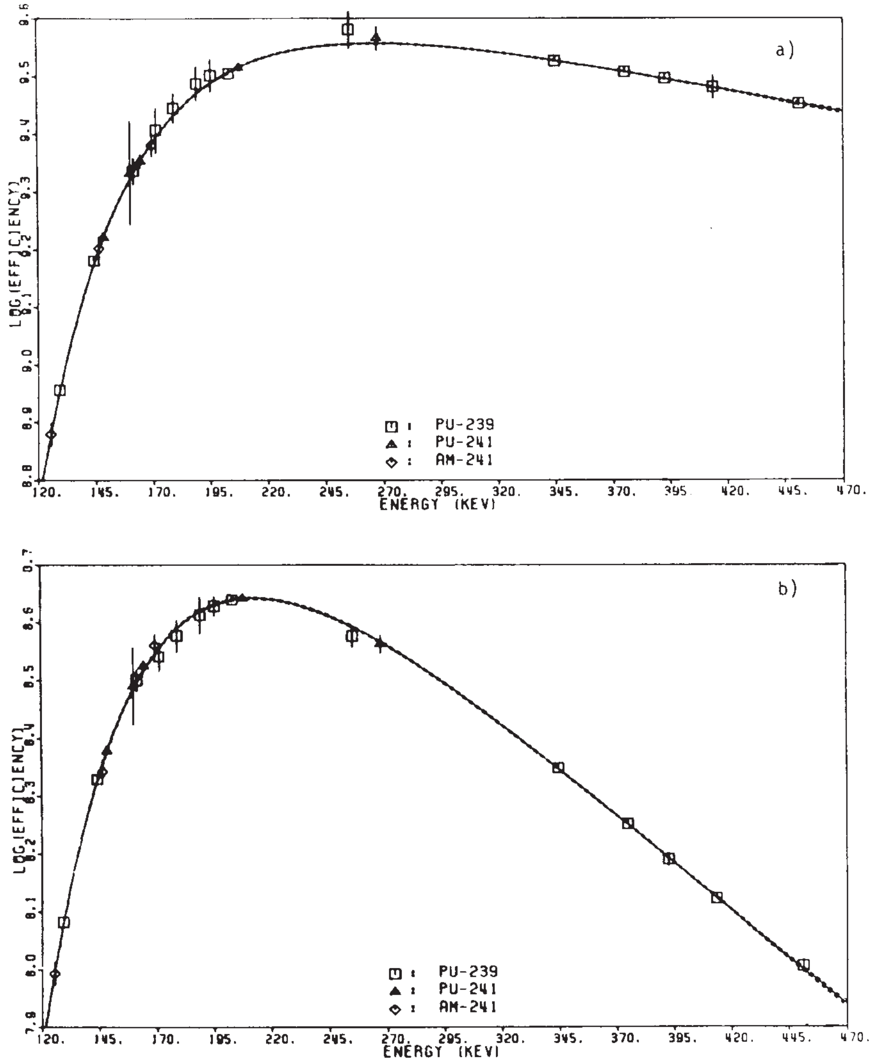
10.2 The half-lives for the various plutonium isotopes and for americium to be used in Eq 1 are listed in Table 3. The gamma-ray branching intensities to be used can be found in Table 2 and are taken from Reference **17.**

10.3 The relative detection efficiencies can be determined through an intrinsic calibration technique. The observed relative gamma-ray intensities and known gamma-ray branching intensities for a particular isotope can be fitted to a functional form of the efficiency-energy relationship by the method of least squares. Energy-intensity data from more than one isotope can be used to improve the fit over the energy range involved. Normalization of these data to the initial set requires that one additional degree of freedom be added to the fitting process for each additional isotope. Fig. 1 shows typical efficiency response curves for PuO_2 samples differing in mass, where data points from ^{239}Pu, ^{241}Pu, and ^{241}Am have been used to obtain the fit.

10.4 Recommended peak pairs to be used for the isotopic abundance ratios are listed in Table 4along with the necessary conversion factors, C_{ij}.

10.5 The beta decay of the ^{241}Pu daughter, ^{237}U, populates levels in ^{237}Np as does the alpha-decay of ^{241}Am. Therefore, some of the gamma-ray transitions used for ^{241}Pu atom ratio determinations must be corrected for the amount of americium feeding. In order to account for this interference, Eq 1 is modified as follows:

$$\frac{N_{241}}{N_{239}} = C_{ij}\,\frac{I_i^{241}}{\epsilon_i}\cdot\frac{\epsilon_j}{I_j^{239}} - C_k\,\frac{N_{Am}}{N_{239}} \qquad (3)$$



NOTE 1—(a)—Typical relative efficiency curve for 2 kg aged plutonium-oxide sample. Relative spectral intensities for ^{239}Pu, ^{241}Pu, and ^{241}Am are used to generate response function. Vertical scale is natural logarithm of relative units. A300 mm^2 by 7 mm planar Ge detector was used for the spectral measurements.

NOTE 2—(b)—Relative efficiency curve for 5 g aliquot of 2 kg sample shown in Note (a).

FIG. 1 Relative Detection Efficiency As a Function of Gamma-Ray Energy

TABLE 4 Recommended Peak Pairs for Determining Isotopic Atom Ratios

Atom Ratio N_i/N_j	Peak Energies E_i, E_j(keV)	C_{ij}[A]	C_k[A]
^{238}Pu/^{241}Pu	152.68, 148.57	1.220	...
^{240}Pu/^{241}Pu	160.31, 148.57	212.75	...
^{240}Pu/^{239}Pu	642.48,[B] 645.97[B]	0.3254	...
^{241}Pu/^{239}Pu	148.57, 129.29	1.991×10^{-2}	...
	208.00, 203.54	6.456×10^{-4}	0.04906
^{241}Am/^{239}Pu	125.29, 129.29	2.758×10^{-2}	...
	335.44, 345.01	2.026×10^{-2}	0.1456
	368.61, 375.04	0.1301	0.1462
	662.42,[B] 645.97[B]	7.354×10^{-4}	...
	721.99,[B] 717.72[B]	2.513×10^{-4}	...

[A] Calculated using branching intensities and half lives from Table 2 and Table 3 respectively.

[B] Possible interferences due to fission products.

$$C_k = \frac{T^{1/2}{}_{241}}{T^{1/2}{}_{Am}} \cdot \frac{B_i^{Am}}{B_i^{241}} \tag{4}$$

where:

$T^{1/2}$ = isotopic half-life, and

B = gamma-ray branching intensity.

The constants, C_k, are listed in Table 4 where appropriate.

10.6 Conversely, several gamma-ray transitions used for ^{241}Am atom ratio determinations must be corrected for the amount of ^{241}Pu feeding. In this case, Eq 3 and Eq 4 can be used after interchanging the ^{241}Pu superscripts and subscripts with those for ^{241}Am (see Table 2).

11. Precision and Bias

11.1 The precision for an atom ratio measurement is a function of several interrelated factors and, therefore, a single predetermined value cannot be quoted. Each sample analysis must be individually evaluated.

11.2 Major factors that can affect the measurement precision include count rate, count time, absorbers, sample geometry, sample mass, sample isotopic composition, and instrument stability (see Ref **11 and 19.**).

11.2.1 Repeatability improves proportionally with the square root of the count time for a given count rate. Likewise, if pileup effects can be neglected, repeatability improves proportionally with the square root of the count rate for a constant count time. These effects reflect the fact that the repeatability will be a function primarily of the statistical uncertainties associated with the measured peak areas.

11.2.2 Absorbers, in excess of the recommendations in 5.2, will unnecessarily attenuate the peak intensities in the 100 to 200 keV range, therefore, reducing the measurement precision achievable.

11.2.3 Sample geometry and sample mass can produce effects, such that larger mass samples will not always produce higher count rates. For samples with plutonium masses greater than a few grams, the suggested operating count rates (see 9.1) can usually be achieved.

11.2.4 The physical size, density, and chemical composition of the sample and the materials surrounding the sample and detector determine the amount of gamma-ray scattering. Scattered gamma rays increase the background continuum in the 100 to 200 keV region. A smaller peak-to-continuum ratio degrades the statistical precision achievable for peak areas in this region. This effect is most pronounced for high mass samples (greater than a few hundred grams of plutonium). For this reason small samples usually exhibit a larger peak-to-continuum ratio than do larger samples of the same material.

11.2.5 The relative isotopic abundances affect measurement precision. In general, higher burnup material gives improved precision for ^{238}Pu, ^{240}Pu, and ^{241}Pu peaks. The precision for peaks from ^{239}Pu generally decreases as burnup increases.

11.3 The computer program used to reduce the data can propagate the statistical errors in the peak areas to estimate the statistical precision of the final isotopic results. This may enable the precision to be estimated for each individual sample. The precision predictions of the data analysis program shall be verified by making repeated measurements on selected samples that cover the range of interest.

11.4 For a wide range of sample types, isotopic compositions, and sample masses, typical values for the statistical precision achievable in a few hours counting time (1 to 4 h) for the normalized isotopic abundances will generally fall into the following ranges:

Isotope	Relative Standard Deviation, %
^{238}Pu	0.6–10
^{239}Pu	0.1–0.5
^{240}Pu	0.8–5
^{241}Pu	0.2–0.8
^{241}Am	0.2–10

11.5 Biases in the branching ratios (see Table 2) may produce a bias in the calculated atom ratios. Small biases have been identified in some of the commonly used branching ratios **(7, 20, 21)**. Contributions due to biases or imprecision in the half-lives are smaller than the level of measurement precision generally obtained and can usually be ignored.

11.5.1 Biases in the branching ratios, half-lives, peak areas, and relative efficiency curves may be identified by using reference materials. The biases are typically a few percent or less. After corrections are made, bias in the atom ratios can be reduced to less than 1 % and in most cases less than 0.5 %. If no corrections are made, accuracies of no better than 1 to 3 % may be obtained for some of the isotopic atom ratios.

11.5.2 Uncertainties assigned to atom ratio values of certified reference materials should be assessed for their effect on the overall measurement accuracy of the isotopic system, when these reference materials are used for determining the atom ratio conversion factors.

12. Keywords

12.1 americium-241; calorimetry; gamma-ray spectrometry; isotopics; neutron counting; nondestructive assay; plutonium; special nuclear material; uranium

REFERENCES

(**1**) Gunnink, R., "Use of Isotope Correlation Techniques to Determine $_{242}$Pu Abundance," *Nuclear Materials Management*, Vol 9, No. 2, 1980, pp. 83–93.

(**2**) Dragnev, T. N., "Intrinsic Self-Calibration of Non-Destructive Gamma Spectrometric Measurements—Determination of U, Pu, and ^{241}Am Isotopic Ratios," *Journal of Radioanalytical Chemistry*, Vol 36, 1977, pp. 491–508.

(**3**) M.J., Koskelo, C. G., Wilkins and Fleisner, J.G., "Comparison of the Performance of Different Pltuonium Isotopic Codes Using a Range of Detector Types,"*Proceedings of the 23rd ESARDA Annaul Symposium on Safegaurds and Nuclear Material Management*,Brugge, Belgium, May 7–11, 2001.

(**4**) Sampson, Thomas E., "Gamma-Ray Isotopic Analysis Development at Los Alamos," "Los Alamos National Laboratory Report, LA-13667–MS, November 1999.

(**5**) Fleissner, J. G., Hyman, P.A., "Implications of Pu Isotopic Measurements for "Mixtures" of Differing Isotopic Source Material," *Proceedings of the 43rd Annual Meeting of the Institute of Nuclear Materials Management*, June 2002, Orlando, FL.

(**6**) R. Gunnink, et. al., "MGAU: A New Analysis Code for Measurement of U-235 Enrichments in Arbitrary Samples," IAEA Symposium on INternational Safegaurds, Wienna, Austria, March 8–14, (1994).

(**7**) Fleissner, J. G., Lemming, J.F., and Jarvis, J.Y., "Study of a Two-Detector Method for Measuring Plutonium Isotopics,"*Measuremetn Technology for Safegaurds and Materials Control*, National Institute of Standards and Technology Special Publication 582, June 1980, pp. 555–567.

(**8**) Fleissner, J. G. et. al., "TRIFID, A Second Generation Plutonium Isotopic Analysis System,"*Proceedings of the 30th Annual Meeting of the Institute of Nuclear Materials Management*, July 1989, Orlando, FL, pp. 814–820.

(**9**) Thomas E. Sampson, George W. Nelson, Thomas A. Kelley, "FRAM: A Versatile Code for Analyzing the Isotopic Composition of Plutonium from Gamma-Ray Pulse Height Spectra,"*Los Alamos National Laboratory Report LA-11720–MS*, December 1989.

(**10**) R. Gunnink, "MGA: A Gamma-Ray Spectrum Analysis Code for Determining Plutonium Isotopic Abundances, Volume 1, Methods and Algorithms," UCRL-LR-103220, Vol 1, April 1990.

(**11**) Sampson, T. E., "Plutonium Isotopic Composition by Gamma-Ray Spectroscopy: A Review," LA-10750-MS, Los Alamos National Laboratory, 1986.

(**12**) American Standards Association Sectional Committee N6 and American Nuclear Society Standards Committee, *Nuclear Safety Guide*, USAEC Report TID-7016, Goodyear Atomic Corp., 1961.

(**13**) Metz, C. F., "Analytical Chemistry Laboratories for the Handling of Plutonium" in *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958*, United Nations, New York, NY, Vol 17, 1959, pp. 681–690.

(**14**) Wick, O. J., editor, *Plutonium Handbook*, Vol 2, Gordon and Breach Science Publishers, New York, NY, 1967.

(**15**) Fleissner, J. G., "Nondestructive Assay of Plutonium in Isotopically Heterogeneous Salt Residues," *ANS-INMM Safeguards Technology Conference*, Hilton Head, SC, November 28, 1983, MLM-3124 (OP), Mound Laboratories, Miamisburg, OH.

(**16**) Fleissner, J. G., and Hume, M. W., "Comparison of Destructive and Nondestructive Assay of Heterogeneous Salt Residues," RFP-3876, Rockwell International, Rocky Flats Plant, March 1986.

(**17**) Gunnink, R., Evans, J. E., and Prindle, A. L., "A Re-Evaluation of the Gamma-Ray Energies and Absolute Branching Intensities of ^{237}U, $_{238}$Pu, and ^{241}Am," UCRL-52139, Lawrence Livermore National Laboratory, 1976.

(**18**) Sampson, T. E., and Gunnink, R., "The Propagation of Errors in the Measurement of Plutonium Isotopic Composition by Gamma-Ray Spectroscopy," *Journal of the Institute of Nuclear Materials Management*, Vol XII, No. 2, Summer 1983.

(**19**) Fleissner, J. G., Oertel, C. P., and Garrett, A. G., "A High Count Rate Gamma-Ray Spectrometer System for Plutonium Isotopic Measurements," *Nuclear Materials Management*, Vol 14 (Proceedings Issue), 1985, pp. 45–56.

(**20**) Ottmar, H., "Results from an Interlaboratory Exercise on the Determination of Plutonium Isotopic Ratios by Gamma Spectrometry," KfK 3149 (ESARDA 1/81), Kernforschungszentrum Karlsruhe, 1981.

(**21**) Bingham, C. B., and McGonnagle, W. J.," A Critical Comparison of Mass- and Gamma-Ray Spectrometric Measurements of Plutonium Isotopic Reference Materials," *Nuclear Materials Management*, Vol XIV, 1985, pp. 320–324.