



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

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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 ²⁴²Pu 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 % ²³⁹Pu. 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.*

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.

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)O₂)

C982 Guide for Selecting Components for Energy-Dispersive X-Ray Fluorescence (XRF) Systems (Withdrawn 2008)⁴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 ²⁵²Cf Shuffler

C1458 Test Method for Nondestructive Assay of Plutonium, Tritium and ²⁴¹Am 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

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:

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

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.

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

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 $^{241}\text{Am}/^{239}\text{Pu}$ 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 $^{241}\text{Am}/^{239}\text{Pu}$ 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 ²⁴¹Am concentrations. Random summing of the intense 59.5-keV ²⁴¹Am 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 (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 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 ²⁴¹Pu is shown in Fig. 1. The alpha decay branch proceeds through the daughter ²³⁷U which decays with a 6.75 day half-life to ²³⁷Np. 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 ²³⁷U may be used to directly determine ²⁴¹Pu. These major gamma rays from the decay of ²³⁷U also have an identical energy component from the beta decay branch of ²⁴¹Pu proceeding through ²⁴¹Am. The ²⁴¹Am 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 ²⁴¹Pu to ²³⁷U must be used to determine the ²⁴¹Pu isotopic fraction. At all times the gamma rays from the decay of ²³⁷U 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 non-uniform with no adverse effect on the results.

7.2.5 The ²⁴¹Am/²³⁹Pu atom ratio must be uniform in all the plutonium in the item, in order to obtain reliable specific power

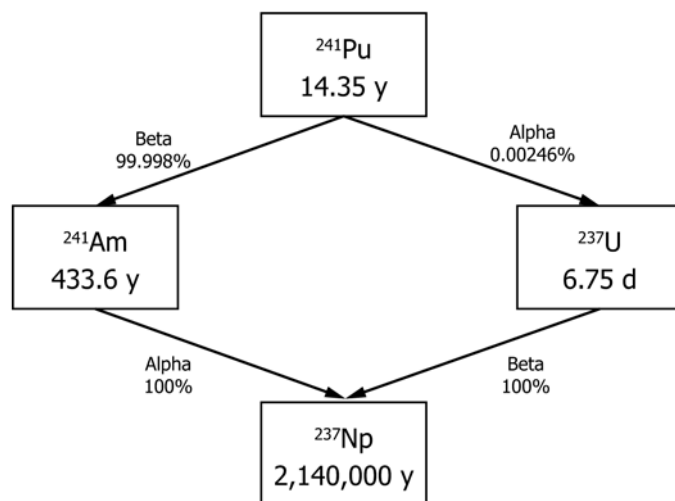


FIG. 1 The Decay of ²⁴¹Pu

measurements to use in interpreting calorimetry results. Certain types of Pu materials with non-homogeneous Am-Pu distributions (salt residues) have been shown to be amenable to assay by this test method with slight modifications (16, 17). 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 those with strong (alpha, n) neutron emissions, should not be stored in the vicinity of the HPGe detector. High energy neutrons emitted by these materials can produce trapping centers in the HPGe crystal and severely degrade the resolution and peak shape of the detector. The use of 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-ray-emitting check source or a plutonium-bearing item because the plutonium gamma-ray energies are well known. A listing of the principal gamma rays emitted from plutonium is given in Table 2. See also Test Methods E181 and Refs (7) and (18).

8.2 Reference Materials:

8.2.1 The expression relating atom ratios to detected peak intensities contains only fundamental constants (see 3.5) and does not depend upon reference standards. Reference standards can be used to identify biases in the values of measured results and as an aid in identifying possible spectral interferences.

8.2.2 Working reference materials with isotopic composition traceable to the National Measurement System 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 and biases. Working reference materials traceable to the National Measurement System should be prepared and validated by other analysis techniques (see Test Methods C697, C698, and E267).

8.3 Measurement Control:

TABLE 2 Energies and Gamma-Ray Branching Intensities^A of Principal Pu and Am Spectral Peaks

NOTE 1—The Branching Intensity for ²⁴¹Pu–²³⁷U gamma rays includes the 2.45×10^{-5} branching fraction for the alpha decay of ²⁴¹Pu to ²³⁷U. See Fig. 2.

Isotope	Energy (keV)	Branching Intensity (γ/disintegration, %)
²⁴¹ Am	59.54	35.9
²⁴⁰ Pu	104.23	7.08×10^{-3}
²⁴¹ Am	125.30	4.08×10^{-3}
²³⁹ Pu	129.30	6.31×10^{-3}
²⁴¹ Pu	148.57	1.85×10^{-4}
²³⁸ Pu	152.72	9.37×10^{-4}
²⁴⁰ Pu	160.31	4.02×10^{-4}
²⁴¹ Pu– ²³⁷ U	164.61	4.53×10^{-5}
²⁴¹ Am	164.61	6.67×10^{-5}
²³⁹ Pu	203.55	5.69×10^{-4}
²⁴¹ Pu– ²³⁷ U	208.00	5.18×10^{-4}
²⁴¹ Am	208.00	7.91×10^{-4}
²⁴¹ Pu– ²³⁷ U	332.36	2.93×10^{-5}
²⁴¹ Am	332.36	1.49×10^{-4}
²⁴¹ Pu– ²³⁷ U	335.38	2.33×10^{-6}
²⁴¹ Am	335.38	4.96×10^{-4}
²³⁹ Pu	345.01	5.56×10^{-4}
²³⁹ Pu	375.05	1.55×10^{-3}
²³⁹ Pu	413.71	1.47×10^{-3}
²³⁹ Pu	451.48	1.89×10^{-4}
²⁴⁰ Pu	642.35	1.3×10^{-5}
²³⁹ Pu	645.94	1.52×10^{-5}
²⁴¹ Am	662.40	3.64×10^{-4}
²⁴¹ Am	772.01	1.96×10^{-4}
²³⁸ Pu	766.39	2.2×10^{-5}

^A Energies and branching intensities from Ref (7).

8.3.1 A measurement control program (ANSI N15.36) shall be established in order to identify anomalous measurement results that may be due to instrument failure or operator (procedural) error. 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 item type 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,

TABLE 3 Pu and Am Half-Lives^A

Nuclide	Half-Life (Years)
²³⁸ Pu	87.74 ± 0.04
²³⁹ Pu	24 119 ± 26
²⁴⁰ Pu	6564 ± 11
²⁴¹ Pu	14.348 ± 0.022
²⁴² Pu	376 300 ± 900
²⁴¹ Am	433.6 ± 1.4

^A Half-lives from Test Method C1458.

and counting geometry. To assure the quality of the collected data and analysis methods, the isotopic measurement control program would employ both internal and external checks discussed below.

8.3.3 Internal checks utilize parameters or measurement results from the spectral data of the item being assayed. An important internal check that provides a good indication of the overall hardware performance is the system resolution. System resolution should be monitored on a spectrum-to-spectrum basis using a strong, clean single peak in the spectrum. Another internal check is to monitor the position of certain spectral peaks to identify possible gain shifts. Monitoring the consistency of the isotopic ratios obtained from several peaks from the same isotope can identify possible interferences or inconsistent peak fitting results. 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 working reference materials can be used to verify that the measurement system is still in control. Measurements of the same item on parallel instruments can also be used as a measurement control indicator. Other external techniques are: comparisons of the gamma-ray results from an item to destructive analysis results, participation in interlaboratory exchange programs, comparisons of the present data with historical or stream average data, and the reanalysis of items 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 item-by-item 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.

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 ²⁴¹Am usually produces a substantial contribution to the system dead time; its intensity can be reduced through the use of a Cd or Sn absorber (see 5.2).

9.2 Acquire the spectrum for the length of time necessary to achieve the desired level of statistical 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 may vary from 10 min to over 4 hr depending upon the purpose of the measurement and the plutonium mass in the measured item. Analysis may proceed directly upon the acquired data or the data may be stored on disk for analysis at a later time.

9.3 Analyze the spectral data. The procedural details of the analysis depend upon the specific software (9, 10, 11, 12, 13, 14, 15) used for the analysis.

9.4 The results of an isotopic measurement shall include an uncertainty assigned to each isotopic result. The uncertainties ascribed to the isotopic fractions and isotopic ratios shall be propagated from the statistical uncertainties of the measured peak areas and any uncertainties due to the peak area determination process. Some analysis procedures may also incorporate systematic measurement uncertainties into the final quoted uncertainty. It is incumbent upon the user to understand the components and contributions incorporated in the quoted uncertainty. The description of the included uncertainty components should be found in the user's manual for the software application used.

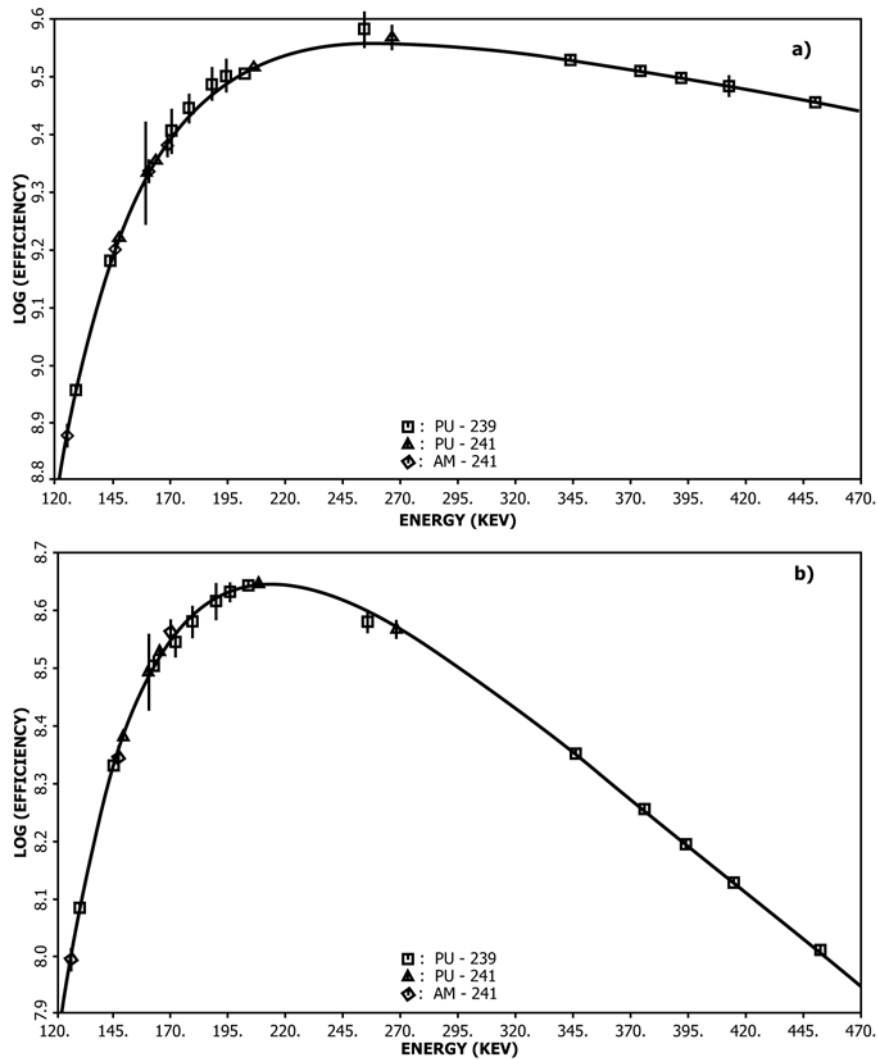
10. Calculation

10.1 The fundamental expression for the isotopic abundance ratio of two isotopes is displayed in Eq 1. All computer codes for gamma ray-isotopic analysis use this fundamental expression although the details of its implementation may vary significantly between codes. Solution of a series of linear equations for each peak area in terms of the isotope activity, branching ratio, and relative efficiency (9, 12, 15) obviates the need to analyze closely-spaced peak pairs and allows the use of all significant gamma rays in the spectrum.

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 for the principal plutonium and ²⁴¹Am gamma-ray peaks most often analyzed are found in Table 2 and are taken from Ref (7). An equally valid set of branching intensities may be found in Ref (18). The differences between the two sets are usually not significant.

10.3 The relative detection efficiencies can be determined through an intrinsic calibration technique, according to Eq 2. The observed experimental relative efficiency values 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. 2 shows typical efficiency response curves for PuO₂ items differing in mass, where data points from ²³⁹Pu, ²⁴¹Pu, and ²⁴¹Am have been used to obtain the fit.

10.4 An empirical polynomial form of the relative efficiency curve (10, 11) has been shown to work well over its range of definition above 120 keV. A physics-based relative efficiency



NOTE 1—(a)—Typical relative efficiency curve for 2 kg aged plutonium-oxide sample over the energy range from 120 to 450 keV. Vertical scale is natural logarithm of relative units. A 300 mm² by 7 mm planar Ge detector was used for the spectral measurements.

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

FIG. 2 Relative Detection Efficiency as a Function of Gamma-Ray Energy

curve (9, 12) can extend the relative efficiency curve across the plutonium K edge at 121.79 keV to enable accurate analyses in the region around 100 keV. A physical relative efficiency curve also eases the problem of extrapolation outside the range of definition, although this must still be done with great care.

10.5 The special issues regarding the decay of ²⁴¹Pu are illustrated in Fig. 1 and have been discussed in 7.2.2. Corrections must be made for the ²⁴¹Am contribution to the “co-energetic” ²⁴¹Pu-²³⁷U peaks which are usually among the most prominent peaks and are usually used (Fig. 2) to help define the relative efficiency curve.

11. Precision and Bias

11.1 Precision:

11.1.1 The precision for gamma-ray isotopic analysis measurement is a function of numerous interrelated factors and, therefore, a single predetermined value cannot be quoted. The analysis of each item must be individually evaluated.

11.1.2 Major factors that can affect the measurement precision include count rate, count time, electronic settings affecting system throughput, absorbers, item geometry, item mass, item isotopic composition, item matrix properties, detector type, and energy range of the analysis (9, 19).

11.1.2.1 Repeatability improves proportionally with the square root of the count time for a given count rate. Likewise, when operating on the linear portion of the count rate-throughput curve, 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.

11.1.2.2 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.

11.1.2.3 The geometry and mass of the measured item can produce effects such that larger plutonium masses will not always produce higher count rates. Reasonable operating count rates can usually be achieved for items with plutonium masses greater than a few grams given a favorable measurement geometry.

11.1.2.4 The physical size, density, and chemical composition of the measured item and the materials surrounding the item 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 items (greater than a few hundred grams of plutonium). For this reason, items with small plutonium mass usually exhibit a larger peak-to-continuum ratio than do larger items of the same material.

11.1.2.5 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.

11.1.2.6 The energy range of the analysis coupled with the type of detector used greatly influences the measurement precision. The measurement precision for the ^{240}Pu isotope is generally the controlling factor in the overall precision of the isotopic distribution measurement. The ^{240}Pu isotope has only three measurable gamma rays for aged plutonium: 104.23 keV, 160.31 keV, and 642.35 keV. The branching intensity decreases approximately a factor of ten for each increase in energy above 104 keV (see Table 2). The complex X-ray region around 100 keV requires a detector with the best possible resolution (see 6.1). A larger, more efficient detector is required for analyses incorporating the 642 keV peak although, the detector energy resolution requirements are less stringent. For measurement items in heavy-walled containers or containers with internal or external shielding the intense lower energy regions of the spectrum may not be available and the analysis for ^{240}Pu has to be carried out at 642 keV. Under many conditions the analysis for ^{240}Pu at 642 keV can produce better precision than analysis of the more intense, lower energy ^{240}Pu gamma rays (9).

11.1.3 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 measurement. The precision predictions of the data analysis program shall be verified by making repeated measurements on selected items covering the range of interest.

11.1.4 For a wide range of item types, isotopic compositions, plutonium masses, and analysis in the 120 to 250 keV region, typical values for the statistical precision achiev-

able in a few hours counting time (1 to 4 h) for the normalized isotopic abundances will generally fall into the ranges displayed in Table 4.

11.1.4.1 Precision values for analysis in the 100 keV region are usually better than in Table 4, when this region is accessible for analysis and the measurement system is appropriate for the analysis.

11.2 Bias:

11.2.1 There are several possible sources of bias in a gamma ray isotopic measurement. The matrix composition of the measured item, nuclear data uncertainties, and radioactive impurities can all lead to biased measurement results.

11.2.2 Contributions to biases from imprecision in the half-lives are smaller than the level of measurement precision generally obtained and can usually be ignored.

11.2.3 The matrix composition of the measured item and the immediate surroundings of the measurement system affect scattering leading to possible distortions in the gamma-ray spectrum. These distortions, as they affect the peak area determination, may give rise to item-dependent biases.

11.2.4 Degradation of the resolution of the HPGe detector usually leads to an increase of measurement bias.

11.2.5 Biases and uncertainties in the branching intensities can directly lead to biased isotopic results. Refs (7) and (18) are two widely-used references for branching intensities. These two references are not independent but where they differ, the differences are typically found to be less than a few percent.

11.2.6 The cumulative effect of these bias sources may be identified and quantified from measurements of reference materials. For these purposes reference materials may be defined as any material with a traceable mass spectrometry measurement because the uncertainties associated with mass spectrometry are, in most cases, significantly less than the uncertainty of the gamma isotopic measurement.

11.2.7 The developers of the isotopic analysis codes have used various methods to account for biases discovered from reference material measurements. Corrections can be made to the branching intensities if it is felt that these are in error. Corrections of several percent are easily justified from the magnitude of the published uncertainties and the magnitude of agreement between the two references previously cited. Corrections to branching intensities or the incorporation of additional multiplicative correction factors are also justified in some cases as correcting for imperfections in the peak fitting and relative efficiency models and fits that do not appear to be random.

TABLE 4 Range of Values of Precision for Measurements in 120 to 450 keV Range

Isotope	Relative Standard Deviation (%)
^{238}Pu	0.6–10
^{239}Pu	0.1–0.5
^{240}Pu	0.5–6
^{241}Pu	0.2–0.8
^{241}Am	0.2–10

11.2.8 The available analysis codes vary in the degree of transparency of these corrections to the user and the accessibility of the corrections to user editing. If the user does have the ability to change or modify correction factors or branching intensities, the user must keep careful records and fully document and test the changes to satisfy local quality assurance requirements.

11.2.9 Some residual bias will remain even after the application of correction factors and “fine tuning” of branching intensities. These residual biases are characterized as item and detector-dependent effects, that is, they arise when considering

a large number of measurements on many different types of items using different detectors over a long period of time. Years of isotopic measurement experience, confirmed by Ref (20), indicate that item and detector dependent biases may be on the order of 1 % (relative) or less for all of the measured isotopes.

12. Keywords

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

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