Designation: C1415 – 14

Standard Test Method for ^{238}Pu Isotopic Abundance By Alpha Spectrometry[1]

1. Scope

1.1 This test method covers the use of alpha spectrometry for determining the ^{238}Pu isotopic abundance in plutonium samples. It is particularly useful for samples in which the ^{238}Pu content is less than 1 % of the total plutonium content. For such samples, mass spectrometric results are vulnerable to bias because of potential interference from any ^{238}U isobar remaining after ion exchange.

1.2 The values stated in SI units are to be regarded as the standard. The values given in parentheses are for information only.

1.3 *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:*[2]

C697 Test Methods for Chemical, Mass Spectrometric, and Spectrochemical Analysis of Nuclear-Grade Plutonium Dioxide Powders and Pellets
C859 Terminology Relating to Nuclear Materials
C1168 Practice for Preparation and Dissolution of Plutonium Materials for Analysis
C1625 Test Method for Uranium and Plutonium Concentrations and Isotopic Abundances by Thermal Ionization Mass Spectrometry
C1672 Test Method for Determination of Uranium or Plutonium Isotopic Composition or Concentration by the Total Evaporation Method Using a Thermal Ionization Mass Spectrometer
D1193 Specification for Reagent Water

3. Terminology

3.1 For definitions of pertinent terms not listed here, see Terminology C859.

4. Summary of Test Method

4.1 This determination method involves the measurement of the alpha-activity ratio of ^{238}Pu and ^{239} Pu + ^{240}Pu. The isotopic analysis of plutonium for the ^{238}Pu isotope requires the prior separation of potentially interfering species. After dissolution of the sample (see Practice C1168), the plutonium is separated from interferences by an anion-exchange purification technique. Nitric acid (HNO_3) is used to adsorb the plutonium fraction on the resin. Interfering ions, principally uranium and americium, are not adsorbed. The plutonium is then eluted with dilute HNO_3 (0.1 *M*). Because an alpha-activity ratio is used, quantitative recovery of the plutonium is not required. The alpha spectrum in the 5 to 6-MeV region is obtained. The total counts in the ^{238}Pu and the ^{239}Pu + ^{240}Pu peaks are obtained and corrected for background. The ^{238}Pu abundance is calculated from the ratio of the alpha activity due to ^{238}Pu and that due to ^{239}Pu + ^{240}Pu. The abundance of ^{239}Pu and ^{240}Pu is determined by mass spectrometry (see Test Methods C697, C1625, or C1672) on a separate portion of the purified sample.

5. Significance and Use

5.1 This test method is used when the determination of ^{238}Pu isotopic abundance is required for plutonium samples.

6. Interferences

6.1 Am-241 is always present as a result of ^{241}Pu decay and is a direct interference that must be removed prior to the determination of ^{238}Pu. The very small amount of ^{230}Th and ^{232}Th which could be present causes insignificant interference with the determination of ^{238}Pu at the level of uncertainty of this test method. Other nuclides that could interfere, such as ^{238}U, ^{243}Am, ^{245}Cm, and ^{249}Bk, are removed by the anion-exchange separation. Any residual uranium, while it does not directly interfere with the alpha-pulse height determination, can raise the salt content of the sample. A high salt content can decrease the resolution of the alpha spectra, and, consequently, decrease the accuracy of the test method.

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[1] This test method is under the jurisdiction of ASTM Committee C26 on Nuclear Fuel Cycle and is the direct responsibility of Subcommittee C26.05 on Methods of Test.

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[2] 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.

7. Apparatus

7.1 *Counting disks of polished platinum, tantalum, or stainless steel,* sized to fit the detection chamber. A disk 25 mm in diameter and 0.5 mm thick has been found to be acceptable.

7.2 *Alpha spectrometer.* This instrument may consist of the following individual components but more typically as an integrated system that is readily interfaced to a computer:

7.2.1 *Silicon surface barrier detector,* with an active area of at least 100 mm^2, a depletion depth of greater than 100 µm, and a resolution of 30 keV or less full width at half maximum (FWHM) (for ^{241}Am 5.486 MeV alpha).

7.2.2 *Evacuable, light-tight chamber* in which the detector and the counting plate on its support can be mounted.

7.2.3 *Preamplifier (charge-sensitive field-effect transistor)* with noise less than 4.6 keV when used with above detector (100 pF capacitance).

7.2.4 *Detector bias supply,* 0 to 150 V, continuously variable, well-regulated and stable, with noise and ripple less than 0.0002 %.

7.2.5 *Main spectroscopy amplifier,* low noise, with variable shaping constants and baseline restoration.

7.2.6 *Biased amplifier and pulse stretcher,* with continuously adjustable post-gain and automatic pile-up rejection.

7.2.7 *Multichannel pulse-height analyzer.* A multichannel analyzer is most versatile and convenient, since it can be used for the acquisition of data from one to four detectors, simultaneously. Even if only one detector is used, such an analyzer has the advantages that background may be stored in another subgroup and subtracted electronically from the spectrum of interest, and that several spectra can be stored and compared. An analyzer that permits the analyst to set windows around the peaks of interest and perform electronic integration is especially convenient. The analyzer should accept pulses 0 to 10 V and 3 to 6 µs in width and should have a capacity of at least 10^6 counts full scale per channel.

7.3 *Heat lamp.*

7.4 *Bunsen burner,* or similar heat source device.

7.5 *Sample beaker,* 30-mL, borosilicate glass.

7.6 *Bottles or vials.*

8. Reagents and Materials

8.1 *Purity of Reagents*—Reagent grade chemicals are used in all tests and conform to the specifications of the Committee on Analytical Reagents of the American Chemical Society.[3]

8.2 *Purity of Water*—Unless otherwise indicated, references to water shall be understood to mean distilled or deionized water (Specification D1193).

8.3 *Nitric acid (HNO_3).* Concentrated (sp gr 1.42).

8.4 *Nitric acid, 4 M.* Add 250 mL of nitric acid (sp gr 1.42) to <750 mL of water and dilute to 1 L.

8.5 *Nitric acid, 0.1 M.* Add 6.2 mL of nitric acid (sp gr 1.42) to <950 mL of water and dilute to 1 L.

8.6 *Anion-exchange resin and column,*[4] 100-200 mesh, containing quaternary ammonium functional groups.

9. Calibration of the Alpha Spectrometer

9.1 *Initial Calibration*—Set the amplifier so that channel zero is about 4.75 MeV. Use a standard alpha source such as the 5.30 MeV particle of ^{210}Po, or the 5.49 MeV particle of ^{241}Am, for energy calibration. Establish the system gain at some convenient value that will cover the needed energy range.

9.2 *Resolution*—Count the standard source and determine the energy span at half the peak height. A full width half maximum of 30 keV or less is desirable, but a FWHM up to 50 keV can be tolerated.

9.3 *Background*—Obtain a background spectrum with a clean counting disk in the chamber.

9.4 *Frequency of Calibration*—The energy calibration and energy resolution should be checked periodically, to maintain the same operating conditions and to check whether the spectrometer has deteriorated. Daily or similarly frequent background counting is advisable, to ensure that neither the chamber nor the detector has been overly contaminated and that an appropriate background correction is accomplished.

10. Procedure

10.1 Transfer an aliquot of approximately 1 mg Pu from a sample obtained from Practice C1168 or equivalent procedure into a 30-mL beaker and take to dryness on a hot plate.

10.2 Add 3-4 mL of 4 M HNO_3 and take to dryness on a hot plate. Cool to room temperature. Repeat this process two more times.

10.3 Add 15 mL of 4 M HNO_3 to an anion-exchange column to precondition it.

10.4 Dissolve the sample with 2-3 mL 4 M HNO_3 and transfer it to the preconditioned anion-exchange column. Let the effluent drain into another beaker; the effluent will be discarded according to the disposal requirements of the specific site.

10.5 Rinse the sample beaker with 2-3 mL of 4 M HNO_3 and transfer the rinse to the column; allow the rinsate to drain. Repeat this rinse step once more.

10.6 Add 5 mL of 4 M HNO_3 to wash the column and allow the column to drain. This waste will be added to the waste already accumulated in 10.4 and 10.5. Repeat this wash step three more times.

10.7 Place a clean bottle or vial under the column and elute the plutonium from the column with two 5–mL portions of 0.1 M HNO_3. The eluant will be used for both alpha and mass spectrometry.

10.8 Mix eluant and transfer a 10 µL aliquot of this solution to a counting disk. Dry under a heat lamp.

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[3] *Reagent Chemicals, American Chemical Society Specifications*, American Chemical Society, Washington, DC. For suggestions on the testing of reagents not listed by the American Chemical society, see *Analar Standards for Laboratory Chemicals*, BDH Ltd. Poole, Dorset, U.K., and the *United States Pharmacopeia and National Formulary*, U.S. Pharmacopeial Convention, Inc. (USPC), Rockville, MD.

[4] Disposable, prefilled columns packed with AG 1-X 8, available from Bio-Rad, Richmond, CA, have been found to be acceptable.

10.9 Fire the disk by slowly heating it to dull redness in a Bunsen burner or similar heat source device.

10.10 Place the counting disk in a chamber of the alpha spectrometer and evacuate the chamber. Count until sufficient counts are accumulated for desired precision.

10.11 Obtain the total count for a sufficient number of channels to include the ^{238}Pu peaks (5.50 + 5.46 MeV) and the ^{239}Pu + ^{240}Pu peaks (5.10 to 5.17 MeV). To obtain the net counts, subtract the background counts in the same channels, adjusted for the same counting time, from the total counts in the peaks, to obtain the net counts.

10.12 Using the aliquot from the purified sample obtained in 10.7, perform mass spectrometric analysis for ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{242}Pu in accordance with Test Methods C697 (Sections 97–105), C1625, or C1672.

11. Calculation

11.1 Calculate the weight percent of ^{238}Pu as follows:

$$W_8 = \frac{C_8 \left(W_9 A_9 + W_0 A_0 \right)}{A_8 C_9} \qquad (1)$$

where:

W_8 = weight percent of ^{238}Pu,
W_9 = weight percent of ^{239}Pu from mass spectrometry (normalized),
W_0 = weight percent of ^{240}Pu from mass spectrometry (normalized),
A_8 = alpha specific activity of ^{238}Pu,
A_9 = alpha specific activity of ^{239}Pu,
A_0 = alpha specific activity of ^{240}Pu,
C_8 = observed net counts in ^{238}Pu peaks, and
C_9 = observed net counts in ^{239}Pu peaks + ^{240}Pu peaks.

11.2 The specific activity of a nuclide (in disintegrations per minute per microgram) is calculated from its half-life by the formula:

$$\text{Specific activity, d/min/}\mu\text{g} = \frac{7.937 \times 10^{11}}{A_w \; t_{1/2}} \qquad (2)$$

where:

A_w = atomic weight of the nuclide, and
$t_{1/2}$ = half-life years, (see Table 1[5]).

TABLE 1 Principal Alpha Peaks of Interest[A]

Nuclide	$t_{1/2}$, years	Alpha-Particle Energy, MeV	Alpha-Particle Abundances, %
^{238}Pu	87.7 ± 0.3	5.50	70.9
		5.46	29.0
^{239}Pu	24,110 ± 30	5.16	73.3
		5.14	15.1
		5.10	11.5
^{240}Pu	6563 ± 7	5.17	72.8
		5.12	27.1
^{241}Am	432.2 ± 0.7	5.49	85.3
		5.44	13.0
		5.39	1.6

[A] Data taken from S.Y.F. Chu, L.P. Ekstrom, and R.B. Firestone, *The Lund/LBNL Nuclear Data Search*, April 1998.

12. Precision and Bias

12.1 New Brunswick Laboratory Certified Reference Material 137, Plutonium Isotopic Standard, was measured for ^{238}Pu content 36 times by one analyst on 6 different days over a period of 10 months. For each measurement, approximately 10 000 counts were collected in the ^{238}Pu alpha peak. The percent relative difference from the reference value (defined as (observed value - reference value)/reference value (100)), was calculated for each determination. One determination's value was an apparent statistical outlier (its magnitude was over 4 times greater than any of the other 35 values). All results were calculated both with and without the suspected outlier (results without the value are indicated by parenthesis). The mean percent relative difference of the data is 1.26 (0.64) which serves as an estimate of bias. The simple standard deviation of the data is 4.18 (1.94) which, in the absence of statistically significant sources of variation, serves as an estimate of precision. Removing the suspect data point from calculations however, gives a statistically significant day-to-day variation in the data. Variance component techniques[6] give an overall estimate of precision without the outlier of 2.00 which includes a component due to day-to-day variation.

12.2 At the time of analysis, the ^{238}Pu content of CRM 137 constituted 0.25 weight percent of the total plutonium content, with an uncertainty (95 % confidence interval for a single determination) on that content of 0.006 weight percent, or 2.4 % of the value.

13. Keywords

13.1 alpha spectrometry; ion exchange; plutonium–238 abundance

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[5] The constants in Table 1 were used in the calculation of the measured values used to determine precision and bias of the method. Other compilations of these constants may be used as agreed upon between shipper and receiver. The database from which the constants are taken should be stated.

[6] *Statistical Methods for Nuclear Material Management*, W. Michael Bowen and Carl A. Bennett, Eds., NUREG/CR-4604, PNL-5849 (US Nuclear Regulatory Commission, Washington, DC, 1988), pp. 265–269.

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