



Standard Test Method for Quantitative Determination of Americium 241 in Plutonium by Gamma-Ray Spectrometry¹

This standard is issued under the fixed designation C 1268; 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 covers the quantitative determination of americium 241 by gamma-ray spectrometry in plutonium nitrate solution samples that do not contain significant amounts of radioactive fission products or other high specific activity gamma-ray emitters.

1.2 This test method can be used to determine the americium 241 in samples of plutonium metal, oxide and other solid forms, when the solid is appropriately sampled and dissolved.

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:

C 758 Test Methods for Chemical, Mass Spectrometric, Spectrochemical, Nuclear, and Radiochemical Analysis of Nuclear-Grade Plutonium Metal²

C 759 Test Methods for Chemical, Mass Spectrometric, Spectrochemical, Nuclear, and Radiochemical Analysis of Nuclear-Grade Plutonium Nitrate Solutions²

C 859 Terminology Relating to Nuclear Materials²

C 982 Guide for Selecting Components for Energy Dispersive X-ray Fluorescence (XRF) Systems²

C 1009 Guide for Establishing a Quality Assurance Program for Analytical Chemistry Laboratories Within the Nuclear Industry²

C 1168 Practice for Preparation and Dissolution of Plutonium Materials for Analysis²

E 181 Methods for Detector Calibration and Analysis of Radionuclides³

2.2 ANSI Standards:

ANSI N15.20 Guide to Calibrating Nondestructive Assay Systems⁴

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 *Annual Book of ASTM Standards*, Vol 12.01.

³ *Annual Book of ASTM Standards*, Vol 12.02.

⁴ Available from American National Standards Institute, 11 W. 42nd St., 13th Floor, New York, NY 10036.

ANSI N15.35 Guide to Preparing Calibration Material for Nondestructive Assay Systems that Count Passive Gamma Rays⁴

ANSI N15.37 Guide to the Automation of Nondestructive Assay Systems for Nuclear Material Control⁴

2.3 *U.S. Nuclear Regulatory Commission Regulatory Guides:*

Regulatory Guide 5.9, Rev. 2—Guidelines for Germanium Spectroscopy Systems for Measurement of Special Nuclear Materials⁵

Regulatory Guide 5.53, Rev. 1—Qualification, Calibration, and Error Estimation Methods for Nondestructive Assay⁵

3. Summary of Test Method

3.1 An aliquot of the sample that contains about 10 to 100 ng of americium 241 is analyzed by measuring the intensity of the characteristic 59.5 keV gamma ray emitted by americium 241.

3.2 Multiple sample geometries may be used if an appropriate calibration for each geometry is made.

3.3 The sample geometry must be reproducible. This includes the physical characteristics of the sample container, the positioning of the sample, and the volume of sample viewed by the gamma-ray detector.

3.4 Electronic corrections are made, if required, for the effects of pulse pile-up and dead time losses due to the activity of the sample. The necessity of dead time and pulse pile-up corrections can be reduced by sample dilution to control count rates.

3.5 A correction is made for the contribution to the 59.5 keV intensity due to gamma rays produced in the decay of uranium 237.

3.6 The relationship between the measured gamma-ray intensity and the americium 241 content is determined by the use of appropriate standards.

4. Significance and Use

4.1 This test method allows the determination of americium 241 in a plutonium solution without separation of the americium from the plutonium. It is generally applicable to any solution containing americium 241.

⁵ Available from U.S. Nuclear Regulatory Commission, 1717 H ST., NW, Washington, DC 20555.

4.2 The americium 241 in solid plutonium materials may be determined when these materials are dissolved (see Practice C 1168).

4.3 When the plutonium solution contains unacceptable levels of fission products or other materials, this method may be used following a tri-n-octylphosphine oxide (TOPO) extraction, ion exchange or other similar separation techniques (see Test Methods C 758 and C 759).

4.4 This test method is less subject to interferences from plutonium than alpha counting since the energy of the gamma ray used for the analysis is better resolved from other gamma rays than the alpha particle energies used for alpha counting.

4.5 The minimal sample preparation reduces the amount of sample handling and exposure to the analyst.

4.6 This test method is applicable only to homogeneous solutions. This test method is not suitable for solutions containing solids.

4.7 Solutions containing as little as 1×10^{-5} g/L americium 241 may be analyzed using this method. The lower limit depends on the detector used and the counting geometry. Solutions containing high concentrations may be analyzed following an appropriate dilution.

5. Interferences

5.1 The presence of other radioactive nuclides in the sample or in the vicinity of the detector may produce interferences. These may be due to the Compton scattering of high energy gamma rays which contribute to the background in the region of interest or from gamma rays with energies close to the energies used for the analysis.

5.2 The presence of uranium 237 will interfere if a correction is not applied. This interference will lead to an over estimation of the amount of americium 241 present. This interference is especially pronounced in plutonium from which the americium has recently been separated.

5.3 The presence of radioactive materials in the vicinity of the gamma-ray detector which are not in the sample may create interferences if detector shielding is not adequate. These interferences may be due to the Compton scattering of high energy gamma rays which contribute to the background in the region of interest or from gamma rays with energies close to the energies used for the analysis.

6. Apparatus

6.1 *High-Resolution Gamma Ray Counting System*—A high resolution gamma-ray counting system is required. General guidelines for the selection of detectors and signal processing electronics are discussed in Guide C 982 and NRC Regulatory Guide 5.9. Data acquisition systems are addressed in ANSI 15.37 and NRC Regulatory Guide 5.9. This system should include the following items as a minimum.

6.1.1 *Germanium Photon Detector with Integral Preamplifier*—A coaxial type detector should typically have a full width at half maximum resolution of 850 eV or less at 122 keV and 2.0 keV or less at 1332 keV. A planar type detector should typically have a full width at half maximum resolution of 600 eV or less at 122 keV. Consideration should be given to the use of a high efficiency detector to enhance the ability to analyze low levels of americium.

6.1.2 *High Voltage Power Supply*—A high voltage power supply with voltage range and current output compatible with the detector selected is required. It is desirable that the voltage output be continuously adjustable.

6.1.3 *Nuclear Spectroscopy Amplifier*—Select a nuclear spectroscopy amplifier with pulse shaping, baseline restoration, and pulse pile-up rejection circuitry.

6.1.4 *Multichannel Pulse Height Analyzer (MCA)*—Select an MCA with a minimum of 2048 channels. It is desirable that the MCA be compatible with computerized operations so that data acquisition and analysis may be automated. The analog to digital converter (ADC) associated with the MCA should have a clock rate of at least 100 MHz and the capability of digitizing the input voltage range into a minimum of 2048 channels (other types of ADC's which provide equivalent capabilities can be used). The ADC should also have dead time and pulse pile-up correction capabilities.

6.2 *Sample Holder*, incorporating shielding to limit the interferences from background radiation sources, is required. Collimation to restrict the view of the detector to a portion of the sample may be required. The sample holder may incorporate more than one sample position. The sample holder shall provide reproducible positioning for each sample position so that a consistent volume or portion of the sample is viewed by the detector.

6.3 *Sample Vials* of sufficient volume to contain the desired sample as described in 9.2 are required. The sample vials should be made of low density materials and have reproducible dimensions such as wall thickness and internal diameter. Vials with identical dimensions should be used for samples and standards.

7. Hazards

7.1 Plutonium and americium bearing materials are radioactive and toxic. Adequate laboratory facilities, gloved boxes, fume hoods, etc., along with safe techniques must be used in handling samples containing these materials. A detailed discussion of all the precautions necessary is beyond the scope of this test method; however, personnel who handle these materials should be familiar with such safe handling practices.

7.2 Solutions and solids containing radioactive materials represent a potential for high radiation exposure to personnel handling them. Appropriate sample shielding, sample handling procedures, and radiation monitoring should be employed to ensure personnel protection.

8. Calibration and Standardization

8.1 Calibrate the counting system for energy (eV/channel) in the range 0 to 300 keV using a radioactive source or sources which emit gamma rays with well known energies. A plutonium source is an obvious choice. See Methods E 181, ANSI N15.20, and U.S. Regulatory Guide 5.53 for further guidance.

8.2 Determine the relative detection efficiency (counts/emitted gamma ray) of the counting system in the 0 to 300 keV range. Specifically, the efficiency at 59.5 keV and 208 keV needs to be determined. See Methods E 181, ANSI N15.20 and U.S. Regulatory Guide 5.53 for further guidance.

8.3 The relationship between the mass of americium 241 and the number of 59.5 keV gamma rays is established through

fundamental physics and basic nuclear constants, that is, the number of 59.5 keV gamma rays/sec/gram americium 241 = 4.543×10^{10} .

9. Procedure

9.1 If necessary, prepare a plutonium solution from a solid sample following the procedure in Practice C 1168 or other dissolution procedure.

9.2 Determine the amount of solution and the dilution required to provide 10 to 100 ng of americium 241 in the selected sample volume. The sample volume viewed by the detector should be consistent for the samples and standards used, regardless of the concentration.

9.3 Determine the counting time necessary to achieve the desired statistical counting precision. Samples which contain more americium will generally require less time to achieve the same statistical precision.

9.4 Quantitatively transfer the predetermined volume of solution from 9.2 into a sample vial and close.

9.5 Place the vial in the counting system sample holder and acquire a spectrum. The detector should see a consistent portion of the sample volume. The same counting geometry and sample size as used for the standards must be used.

9.6 Record the sample counting time, sample volume, dilution factor, and counting geometry used if more than one is available.

10. Calculation

10.1 Using the same methods as used for the calibration, determine the background corrected net count rates for the 59.5 keV gamma ray and the 208 keV gamma ray using the spectral data acquired in 9.5.

10.2 Calculate the 59.5 keV counting rate due to americium 241 in the sample.

$$R_{Am}(59) = \frac{R_{obs}(59) / D(59) - B_U R_{obs}(208) / D(208)}{1 - B_U / B_{Am}} \quad (1)$$

where:

$R_{Am}(59)$ = 59.5 keV rate (gamma rays/s) due to americium 241,

$R_{obs}(59)$ = measured 59.5 keV rate (counts/s),

$D(59)$ = detection efficiency (counts/gamma ray) at 59.5 keV,

$R_{obs}(208)$ = measured 208 keV rate (counts/s),

$D(208)$ = detection efficiency (counts/gamma ray) at 208 keV,

B_U = 1.5668, and

B_{Am} = 45385.6.

NOTE 1— B_U and B_{Am} are dimensionless constants derived from the half-lives of uranium 237 and americium 241 and the branching ratios of the 59.5 and 208 keV gamma rays. The factor $(1 - B_U / B_{Am})$ may be neglected for most applications.

10.3 Calculate the amount of americium 241 present in the sample using the count rate from 10.2 and the factor in 8.3.

10.4 Using the dilution factor for the sample calculate the amount of americium 241 in the original solution.

11. Measurement Control

11.1 Establish a measurement control program for the analytical method. Section 12 of Guide C 1009 provides

further guidance in this area.

11.2 As a minimum, the following periodic checks should be made.

11.2.1 Make a daily check of all instrument settings and of the energy calibration of the counting system prior to any measurement or series of measurements.

11.2.2 Make a daily measurement of the counting room background. Ideally a measurement of the room background should be made both before and after any series of americium determinations.

11.2.3 Make a daily measurement of an americium standard or sample with a known concentration to provide a measurement bias check.

11.2.4 Make weekly replicate measurements of a standard or sample to determine the precision of the measurement method.

11.3 It is recommended that control charts and other periodic statistical analysis of the precision and bias data be used.

12. Precision and Bias

12.1 The precision of the assay is a function of counting statistics. Precision may be improved with increased counting time.

12.2 Variations in sample vial geometry and positioning will affect the precision of the measurement.

12.3 Differences in the plutonium and acid concentration between the sample and the calibration standards may cause a bias due to self attenuation in the sample.

12.4 The calibration of standard sources, including errors introduced in using a standard radioactive solution or aliquot thereof, to prepare a working standard for bias correction may result in a bias.

12.5 The full energy peak efficiency at a given energy determined from the calibration function may introduce a bias.

12.6 Errors in preparation including sample dilution, sample transfer by pipetting, etc. can result in a bias.

12.7 Samples producing high count rates may cause a bias due to dead time losses and pulse pileup. This bias will be dependent on the adequacy of the corrections provided by the pulse pile-up rejection and dead time correction circuits in the counting electronics. Generally count rates that cause significant problems can be easily avoided by sample dilution. In general the total count rate should be less than 20 000 counts per second to minimize biases.

12.8 One hundred and three replicate analyses of standards performed at a single laboratory (see the appendix) show that the method, when correctly applied, can be free of statistically significant bias.

12.9 Total method precision is strongly dependent on the qualification/training of the personnel performing the measurement. The number of individuals performing the measurement also influences the overall precision. Replicate analyses of standards by 17 different analysts at a single laboratory over a 19-month period (see appendix) have resulted in individual precisions as measured by the percent relative standard deviation (%RSD) from 1.13 % to 7.43 %. The single laboratory precision over all the data is about 3 % to 5 % depending on whether certain assay results are deemed to be outliers or not.

13. Keywords

13.1 americium 241; gamma-ray spectrometry; nondestructive analysis; plutonium solutions

APPENDIX

(Nonmandatory Information)

X1. DATA FOR PLUTONIUM CONCENTRATION

X1.1 Table X1.1 provides data for plutonium concentration.

TABLE X1.1 Data Set^A

NOTE—This data set contains a total of 103 observations made by 17 different individuals over a 19-month period. There are six observations in the data set that may be considered to be statistical outliers. Removing these outliers (indicated by asterisks in the data set) results in 97 observations with an average recovery of 99.66 % and a standard deviation of 3.37 %. Five of the six outliers occurred in a two week period which may indicate a problem with the method during that time. Analysis of the data set also reveals a slight variation which is time and individual dependent.

	Date	Analyst	Standard Value (g Am/L)	Measurement Value (g Am/L)	Difference	Recovery, %
1	01/02/90	69	0.00016577	0.000176	0.0000102	106.1712 %
2	01/08/90	38	0.00016744	0.000170	0.0000026	101.5289 %
3	01/10/90	87	0.00016804	0.000174	0.0000060	103.5468 %
4	01/10/90	69	0.00016812	0.000179	0.0000109	106.4716 %
5	01/16/90	69	0.00016975	0.000169	-0.0000008	99.5582 %
6	01/16/90	38	0.00016984	0.000162	-0.0000078	95.3839 %
7	01/23/90	87	0.00017183	0.000167	-0.0000048	97.1891 %
8	01/24/90	17	0.00017195	0.000173	0.0000010	100.6106 %
9	01/30/90	69	0.00017368	0.000177	0.0000033	101.9116 %
10	02/02/90	38	0.00017460	0.000201	0.0000264	115.1203 %*
11	02/06/90	17	0.00017570	0.000177	0.0000013	100.7399 %
12	02/06/90	70	0.00017576	0.000203	0.0000272	115.4984 %*
13	02/14/90	69	0.00017798	0.000204	0.0000260	114.6196 %*
14	02/14/90	17	0.00017801	0.000207	0.0000290	116.2856 %*
15	02/21/90	87	0.00018002	0.000176	-0.0000040	97.7669 %
16	02/22/90	17	0.00018025	0.000173	-0.0000072	95.9778 %
17	03/01/90	38	0.00018227	0.000186	0.0000037	102.0464 %
18	03/07/90	50	0.00018391	0.000201	0.0000171	109.2926 %
19	03/07/90	17	0.00018393	0.000192	0.0000081	104.3875 %
20	03/07/90	21	0.00018397	0.000180	-0.0000040	97.8420 %
21	03/13/90	51	0.00018564	0.000183	-0.0000026	98.5779 %
22	03/20/90	21	0.00018773	0.000180	-0.0000077	95.8824 %
23	03/21/90	38	0.00018792	0.000181	-0.0000069	96.3176 %
24	03/29/90	38	0.00019021	0.000191	0.0000008	100.4153 %
25	04/03/90	00	0.00020651	0.000224	0.0000175	108.4693 %
26	04/04/90	99	0.00020688	0.000178	-0.0000289	86.0402 %*
27	04/05/90	38	0.00020701	0.000208	0.0000010	100.4782 %
28	04/09/90	51	0.00020831	0.000198	-0.0000103	95.0506 %
29	04/10/90	99	0.00020852	0.000211	0.0000025	101.1893 %
30	04/13/90	99	0.00020956	0.000210	0.0000004	100.2100 %
31	04/18/90	54	0.00021100	0.000226	0.0000150	107.1090 %
32	04/18/90	38	0.00021109	0.000215	0.0000039	101.8523 %
33	04/19/90	87	0.00021116	0.000223	0.0000118	105.6071 %
34	04/25/90	69	0.00021305	0.000204	-0.0000091	95.7522 %
35	04/25/90	99	0.00021313	0.000214	0.0000009	100.4082 %
36	04/26/90	38	0.00021323	0.000206	-0.0000072	96.6093 %
37	04/30/90	54	0.00021459	0.000208	-0.0000066	96.9290 %
38	04/30/90	73	0.00021470	0.000221	0.0000063	102.9343 %
39	05/01/90	99	0.00021480	0.000217	0.0000022	101.0242 %
40	05/08/90	54	0.00021699	0.000228	0.0000110	105.0740 %
41	05/08/90	28	0.00021702	0.000212	-0.0000050	97.6868 %
42	05/09/90	73	0.00021712	0.000212	-0.0000051	97.6419 %
43	05/15/90	50	0.00021916	0.000221	0.0000018	100.8396 %
44	05/16/90	73	0.00021933	0.000215	-0.0000043	98.0258 %
45	05/17/90	38	0.00021951	0.000222	0.0000025	101.1343 %
46	05/23/90	73	0.00022154	0.000224	0.0000025	101.1104 %
47	05/24/90	99	0.00022166	0.000220	-0.0000017	99.2511 %
48	05/25/90	22	0.00022201	0.000221	-0.0000010	99.5451 %
49	05/30/90	22	0.00022361	0.000230	0.0000064	102.8577 %

TABLE X1.1 *Continued*

	Date	Analyst	Standard Value (g Am/L)	Measurement Value (g Am/L)	Difference	Recovery, %
50	05/31/90	73	0.00022372	0.000222	-0.0000017	99.2312 %
51	05/31/90	99	0.00022382	0.000225	0.0000012	100.5272 %
52	06/05/90	50	0.00022546	0.000220	-0.0000055	97.5783 %
53	06/06/90	55	0.00022551	0.000227	0.0000015	100.6607 %
54	06/06/90	73	0.00022563	0.000227	0.0000014	100.6072 %
55	06/11/90	51	0.00022716	0.000222	-0.0000052	97.7285 %
56	06/12/90	99	0.00022734	0.000228	0.0000007	100.2903 %
57	06/12/90	21	0.00022755	0.000221	-0.0000065	97.1215 %
58	07/31/90	55	0.00024216	0.000252	0.0000098	104.0634 %
59	08/02/90	99	0.00024267	0.000239	-0.0000037	98.4877 %
60	09/26/90	22	0.00025915	0.000262	0.0000029	101.0997 %
61	09/26/90	38	0.00025925	0.000244	-0.0000152	94.1176 %
62	10/11/90	22	0.00026358	0.000269	0.0000054	102.0563 %
63	10/11/90	38	0.00026368	0.000257	-0.0000067	97.4666 %
64	10/18/90	50	0.00026570	0.000264	-0.0000017	99.3602 %
65	10/18/90	87	0.00026580	0.000267	0.0000012	100.4515 %
66	10/25/90	54	0.00026783	0.000262	-0.0000058	97.8232 %
67	10/25/90	52	0.00026793	0.000254	-0.0000139	94.8009 %
68	12/12/90	51	0.00028218	0.000271	-0.0000112	96.0380 %
69	12/21/90	39	0.00028487	0.000279	-0.0000059	97.9394 %
70	12/21/90	38	0.00028491	0.000290	0.0000051	101.7865 %
71	01/02/91	39	0.00028848	0.000263	-0.0000255	91.1675 %
72	01/04/91	38	0.00028909	0.000272	-0.0000171	94.0883 %
73	01/08/91	52	0.00029027	0.000270	-0.0000203	93.0168 %
74	01/09/91	87	0.00029060	0.000274	-0.0000166	94.2877 %
75	01/16/91	52	0.00029270	0.000289	-0.0000037	98.7359 %
76	01/17/91	87	0.00029292	0.000292	-0.0000009	99.6859 %
77	02/15/91	54	0.00030163	0.000295	-0.0000066	97.8019 %
78	02/15/91	38	0.00030166	0.000307	0.0000053	101.7702 %
79	05/06/91	50	0.00032559	0.000317	-0.0000086	97.3617 %
80	05/14/91	55	0.00032791	0.000326	-0.0000019	99.4175 %
81	05/14/91	87	0.00032799	0.000301	-0.0000270	91.7711 %
82	05/21/91	50	0.00033004	0.000331	0.0000010	100.2909 %
83	05/23/91	38	0.00033057	0.000324	-0.0000066	98.0125 %
84	05/30/91	22	0.00033274	0.000335	0.0000023	100.6792 %
85	05/30/91	87	0.00033274	0.000337	0.0000043	101.2803 %
86	06/05/91	50	0.00033449	0.000323	-0.0000115	96.5649 %
87	06/05/91	38	0.00033457	0.000336	0.0000014	100.4274 %
88	06/11/91	55	0.00033633	0.000337	0.0000007	100.1992 %
89	06/11/91	87	0.00033639	0.000346	0.0000096	102.8568 %
90	06/17/91	39	0.00033811	0.000334	-0.0000041	98.7844 %
91	06/17/91	38	0.00033819	0.000350	0.0000118	103.4921 %
92	06/24/91	22	0.00034016	0.000344	0.0000038	101.1289 %
93	06/24/91	87	0.00034031	0.000332	-0.0000083	97.5581 %
94	07/02/91	50	0.00034261	0.000298	-0.0000446	86.9794 %*
95	07/03/91	50	0.00034284	0.000337	-0.0000058	98.2966 %
96	07/03/91	52	0.00034294	0.000350	0.0000071	102.0587 %
97	07/11/91	55	0.00034526	0.000354	0.0000087	102.5314 %
98	07/11/91	87	0.00034534	0.000357	0.0000117	103.3764 %
99	07/17/91	50	0.00034709	0.000334	-0.0000131	96.2286 %
100	07/17/91	38	0.00034712	0.000336	-0.0000111	96.7965 %
101	07/23/91	39	0.00034898	0.000349	0.0000000	100.0057 %
102	07/30/91	39	0.00035098	0.000355	0.0000040	101.1454 %
103	07/30/91	38	0.00035102	0.000345	-0.0000060	98.2850 %
Average Recovery						100.0125 %
% Relative Standard Deviation						4.8668 %
95 % Confidence Interval of the Average					High	100.9637 %
					Low	99.0614 %

^AThis data set was collected at the Westinghouse Hanford Company Plutonium Finishing Plant Engineering Laboratory. The plutonium concentration in the standard is approximately 0.5 g/L. These data were provided by R. A. Hamilton.

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