LA-UR -91-3323

LA-UR--91-3323

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SUBMITTED TO American Nuclear Society
4th International Conference on
Facility Operations--Safeguards Interface
Albuquerque, New Mexico
September 29-October 4, 1991
(FULL PAPER)

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PORTABLE GAMMA-RAY HOLDUP AND ATTRIBUTES MEASUREMENTS OF HIGH- AND VARIABLE-BURNUP PLUTONIUM*

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ABSTRACT

High-burnup-plutonium holdup has been assayed quantitatively by low-resolution gamma-ray spectrometry. The assay was calibrated with four plutonium standards representing a range of fuel burnup and ^{241}Am content. Selection of a calibration standard based on its qualitative spectral similarity to gamma-ray spectra of the process material is partially responsible for the success of these holdup measurements. The spectral analysis method is based on the determination of net counts in a single spectral region of interest (ROI). However, the low-resolution gamma-ray assay signal for the high-burnup plutonium includes unknown amounts of contamination from ^{241}Am. For most needs, the range of calibration standards required for this selection procedure is not available. A new low-resolution gamma-ray spectral analysis procedure for assay of ^{239}Pu has been developed. The procedure uses the calculated isotope activity ratios and the measured net counts in three spectral ROIs to evaluate and remove the ^{241}Am contamination from the ^{239}Pu assay signal on a spectrum-by-spectrum basis. The calibration for the new procedure requires only a single plutonium standard. The procedure also provides a measure of the burnup and age attributes of holdup deposits. The new procedure has been demonstrated using portable gamma-ray spectroscopy equipment for a wide range of plutonium standards and has also been applied to the assay of ^{239}Pu holdup in a mixed oxide fuel fabrication facility.

I. INTRODUCTION

Quantities of low-burnup plutonium as holdup in the process equipment of US production facilities have been measured by simple procedures that use low-resolution gamma-ray detectors, typically NaI(Tl) scintillators, coupled to photomultiplier tubes and portable multichannel analyzers.[1] The timely assay of holdup quantities has important safety consequences in connection with criticality limits for special nuclear material (SNM) deposit masses, as well as accountability benefits in providing inventory quantities, and process control benefits with respect to equipment cleanout requirements. The low-burnup plutonium spectra are relatively easy to interpret because of the isotopic uniformity and low americium content of most materials of this type.

Similar measurements of high-burnup plutonium holdup are of interest to the operators of oxide conversion and fuel fabrication facilities, for the reasons stated above, and to international inspection agencies, for verification of operator declarations. Because of high burnup, however, significant americium ingrowth is typical of fuel cycle materials. Interferences from ^{237}U and ^{241}Am gamma rays in the plutonium assay region, usually chosen to include the intense ^{239}Pu gamma rays at 375 and 414 keV, can give rise to biases of 100% or more [depending on the ^{237}U and ^{241}Am fractions and on the chosen energy region-of-interest (ROI)] in holdup assays derived from calibrations performed with a low-burnup plutonium source. The ^{237}U content, in equilibrium with its ^{241}Pu parent, is a function of fuel burnup. The ^{241}Am fraction is a function of both the burnup and age of the material. Typical holdup deposits for high-throughput processes would consist of unknown admixtures of materials representing a wide range of both variables. The biases resulting from interferences can be avoided by performing the same holdup measurements with solid state, high-resolution hyperpure germanium (HPGe) detectors[2] instead of NaI(Tl), but not without sacrificing convenience, portability, simplicity, and economy.

A recent calibration of the new, polyethylene-moderated ^{3}He slab detection systems for assay, by neutron coincidence counting, of high-burnup plutonium holdup in the glove boxes of a fuel fabrication facility provided an opportunity to simultaneously test the low-resolution gamma-ray holdup assay methods applied to this material type. Because of the unknown and variable effects of interferences on the assays obtained from NaI(Tl) spectra using a single ROI technique, the calibration of the gamma-ray holdup assay for these glove-box measurements was performed with four plutonium reference standards representing a range of burnup and age. The calibration that was finally used to quantify the holdup was chosen based on a visual comparison between spectra from actual holdup deposits and those obtained with the variable-burnup and -age reference standards used in the calibration measurements.

The gamma-ray holdup measurements appeared to be successful in that the results agreed reasonably well with the neutron coincidence assays. However, it is time consuming

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and impractical for most users to perform the holdup calibration with multiple plutonium standards that cover a range of burnup and age and to subsequently choose the calibration based on qualitative comparisons of spectra. Therefore, a new approach to analyzing low-resolution gamma-ray spectra of high-burnup plutonium has been developed and tested. The goal of this approach is to provide a calibration for assay of plutonium holdup that requires only one plutonium reference standard but retains the ability to correct for interferences. The ability to determine attributes that verify burnup and age using this new analysis approach was also tested.

Section II of this paper describes the gamma-ray measurements of holdup in the glove boxes and the comparison of these results with the neutron coincidence measurements. Section III discusses the new analysis method and tests the method on variable burnup plutonium reference standards. Section IV presents the results of re-analysis of the glove-box holdup spectra using the new analysis technique.

II. DESCRIPTION OF HOLDUP MEASUREMENTS OF HIGH-BURNUP PLUTONIUM

A. Holdup Measurement Purposes and Method

The new glove-box assay systems (GBASs) quantify holdup of high-burnup plutonium as mixed oxide (MOX) in the automated bulk processing equipment of the Power Reactor and Nuclear Fuel Development Corporation's (PNC's) Plutonium Fuel Production Facility (PFPF) in Japan. The assay for GBAS is based on counting coincident neutrons from spontaneous fission of the effective ^{240}Pu mass to give the total plutonium mass, as described elsewhere.[3] Each GBAS assay gives the quantity of plutonium in an entire (3-m-tall by 3- or 6-m-wide) glove box in the PFPF process. The GBAS calibrations were performed in situ by placing MOX reference materials inside each process glove box and performing the neutron measurements as prescribed for an assay. The background count rates for these calibration measurements, produced by the plutonium holdup within each glove box, were determined by performing the same GBAS measurements on each glove box with the MOX reference materials removed. Once determined, the GBAS calibrations were applied to the calibration background measurements to quantify the plutonium holdup.

Holdup assays using portable gamma-ray equipment were performed on most of the PFPF glove boxes in advance of the neutron calibration measurements to provide an independent verification of the new GBAS results. The gamma-ray assays are inherently less accurate than the neutron assays in this particular holdup situation, primarily because of the uncertainties in estimated corrections for attenuation of measured gamma rays by the bulk processing equipment. However, the measurements provided a first opportunity to test the generalized-geometry method for holdup assay[1,2] in particular, and low-resolution gamma-ray quantitative and attributes measurements in general, on high-burnup plutonium in a processing facility.

The generalized-geometry holdup assay method uses gamma-ray detectors with cylindrical collimators. The assay signal is the net count rate for a gamma-ray peak. Each chosen peak corresponds to the radioactive decay of a specific isotope. The net peak activity is a measure of the isotope quantity. The method requires the selection of measurement geometries to establish the holdup deposit as either a relatively small, centered point (point source), a long and relatively narrow, centered line (line source), or a relatively large area (area source) in the large and circular detector field of view. By spacing adjacent measurements (performed on a line or area source) a distance equal to the full width at the half maximum of the radial detection response at the deposit surface, uniform coverage of the entire length or surface is assured. The line or area source assay gives isotope mass per unit length or area, respectively, at the measurement location. The average assay result for all locations on the line or area multiplied by the length of the line or by the surface area, respectively, gives the total isotope mass. Knowledge of the fraction of the assay isotope is required to obtain the element mass. The assay result is corrected for attenuation of the gamma rays (of well-defined energies) by the equipment.

The generalized-geometry holdup assay is calibrated in two steps. One step measures the radial detection response at the assay energy to obtain the appropriate geometric factors for the calibration. This requires a point source, not necessarily a standard, of the assay gamma ray. The second step measures a point-source standard of the appropriate material to obtain the attenuation-corrected absolute point response per unit mass of isotope at a known distance r_0.

The holdup assay uses the equipment-attenuation-corrected net peak count rate from a holdup deposit measured at the distance r. This count rate is divided by the absolute response per unit mass and by the appropriate geometric factor (unity for point source assays) and multiplied by $(r/r_0)^n$, where n is 2, 1, or 0 for point, line, or area sources, respectively.

B. Holdup Measurement Equipment and Calibration

The generalized-geometry holdup assays at the PFPF were calibrated at Los Alamos. A 2.5-cm-diam by 5-cm-thick, shielded, cylindrically collimated NaI(Tl) detector with a 1.6-mm-thick cadmium filter and a portable multichannel analyzer (PMCA) were used. Two energy ROIs were set up in the PMCA to obtain the calibration data. These were the assay (375 to 450 keV) and the continuum background (475 to 550 keV) ROIs. This conservative choice for the assay ROI eliminates half of the activity from the intense ^{239}Pu gamma ray at 375 keV to minimize the relative interference effects on the assay. An ROI set on the relatively intense 208-keV gamma-ray peak from ^{237}U decay was used to monitor the gain for the possibility of drift.

The radial detection response was measured with a standard point source of low-burnup plutonium. The absolute response was measured with four different standard point sources representing a large burnup range, including the low-burnup source. The spectra obtained with these four standards are shown in Fig. 1(a)–(d), with the assay and continuum background ROIs shaded. The isotopic compositions and plutonium masses of the four calibration standards are included in Table I. The dramatic differences among the four spectra, particularly near and below the assay ROI, are primarily the results of the ^{241}Pu fraction variability with burnup and the ^{241}Am variability with burnup and age.

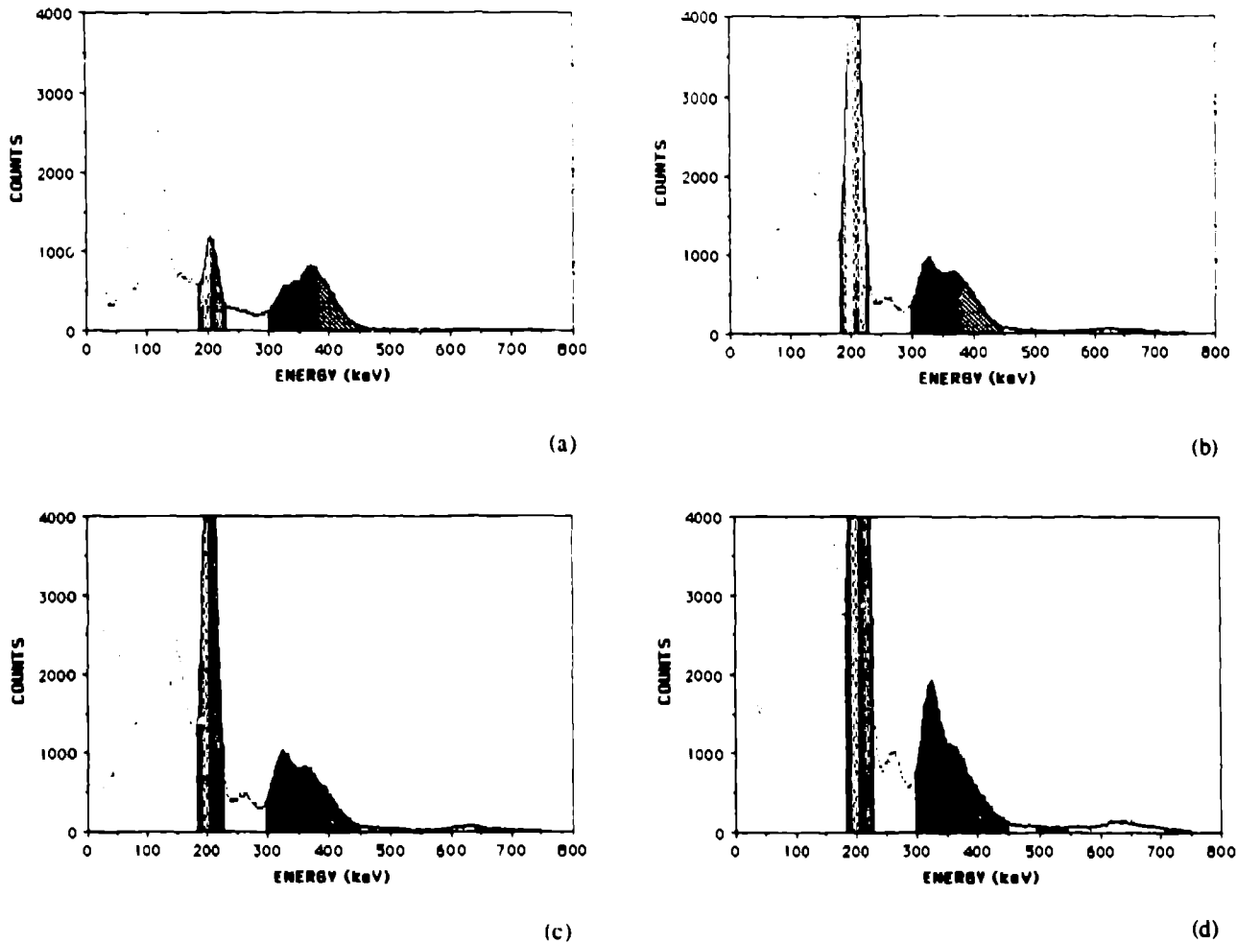


Fig. 1(a)–(d). Low-resolution gamma-ray spectra of four plutonium standards, P1, P4, P5, and P6 (a, b, c, and d, respectively), where P1 is a low-burnup standard and burnup increases with standard number. These four standards were used for the calibration of the original assays of high-burnup-plutonium holdup. The assays were performed using the P6 calibration, based on the similarity between d and the process material spectra. The assay ROI (ROI a) for the original holdup measurements was the 375- to 450-keV shaded ROI. Its continuum background ROI is also shaded. The two lower-energy ROIs (ROI2 at 181–235 keV and ROI3 at 300–375 keV) used in the new (three-ROI) analysis procedure are shaded as well.

C. Holdup Measurement Procedures and Results

Before holdup measurements were performed at the PFPF, two MOX process reference samples, each containing approximately 5 g of plutonium, were measured as point sources. Based on a visual inspection of the spectra obtained from these samples, the calibration obtained with the highest burnup standard point source of the four calibration standards was selected for assaying the process samples for verification purposes. The point source assay results for ^{239}Pu were within 5% of the reference values for these process samples. Subsequently, the holdup assays were performed using the verified calibration (based on the highest-burnup standard).

The gamma-ray holdup measurements were performed at the glove ports of the 3- by 3-m (36-port) and 3- by 6-m (72-port) glove boxes at the PFPF facility. The measurements were performed at one surface (front or back) of each glove box. (The automated process equipment was centered and generally symmetric, front-to-back, within the glove boxes.) The detector was positioned at the glove-port centers, 57 cm from the lengthwise vertical plane through the center of the glove box. A calciner extension to one of the glove boxes was also measured. The spacing between adjacent measurements (in adjacent glove ports) corresponded to the full width at half maximum of the radial detection response in this vertical plane, fulfilling the criterion for uniform coverage. The spectra were digitized into 512 channels

TABLE I. Reference Sample Isotopic Composition and Plutonium Mass

			wt%					
Sample	Updated to	g Pu	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am
P1[a]	4/25/91	0.397	0.011	93.795	5.989	0.170	0.035	0.256
P2	4/25/91	0.397	0.023	89.398	10.103	0.381	0.095	0.330
P3	4/25/91	0.396	0.046	84.707	14.163	0.851	0.233	0.766
P4[a]	4/25/91	0.396	0.105	77.933	19.819	1.572	0.569	1.834
P5[a]	4/25/91	0.396	0.128	76.118	21.282	1.766	0.705	2.041
P6[a]	4/25/91	0.394	0.913	66.883	24.076	4.541	3.587	4.455
P7	4/25/91	0.393	1.235	62.602	25.846	5.592	4.726	4.439
C8	7/21/91	5.856	0.011	93.460	6.314	0.175	0.040	0.152
C9	7/21/91	5.846	0.068	84.532	14.234	0.808	0.359	0.437
C10	7/21/91	5.788	0.823	74.223	18.513	4.338	2.103	2.364
C11	7/21/91	5.771	1.168	63.475	25.782	5.318	4.257	2.913

[a]Used in calibration of original holdup measurements

and were acquired in 50- to 100-s counting periods. All spectra were stored on PMCA magnetic tape.

For each glove box, the net count rates for the assay ROI were averaged for all glove-port locations, and room background was subtracted from the average. The correction factors for equipment attenuation were applied to the average net count rates. These were calculated from the known transmission of 400-keV gamma rays through steel. Each glove box was assigned an effective (estimated) attenuating thickness of 3, 6, or 10 mm of steel, corresponding to correction factors of 1.23, 1.51, or 2.00, respectively, based on a visual inspection of the equipment within the box and on the assumption that the holdup is deposited within the equipment in the glove box. The correction factor of 3 for attenuation of gamma rays by the calciner equipment was based on an estimated steel thickness of 16 mm.

The area-source calibration was applied to the average, attenuation-corrected net count rate for each glove box. The assay result, in g $^{239}Pu/cm^2$, for a given glove box was multiplied by the total deposit surface area (typically 9×10^4 cm^2) to give the mass (in grams) of ^{239}Pu holdup in the glove box. The line source calibration was applied to the calciner average (attenuation-corrected net) count rate. The assay result, in g $^{239}Pu/cm$, was multiplied by the total calciner length (200 cm) to give the mass (in grams) of ^{239}Pu holdup in the calciner. The ^{239}Pu mass for each glove box (and for the calciner) was divided by the operator-declared stream-average weight fraction of the ^{239}Pu isotope to yield the gamma-ray assay of plutonium holdup for each glove box.

D. Comparison of Neutron and Gamma-Ray Holdup Results

Table II gives the average, attenuation-corrected net count rate in the assay ROI (R_a, s^{-1}) and its relative uncertainty (determined from counting statistics) for each glove box. The steel attenuation correction factors for the assay ROI and the operator-declared ^{239}Pu weight fractions are also given. The plutonium holdup assays obtained with these R_a values were compared with the holdup assays obtained from neutron coincidence counting. The neutron assays were obtained for combined (wall and aisle) halves of the double-width glove boxes. These results were compared with the combined gamma-ray assays for the two halves. The comparison results are given in Table II. The average assay ratio (gamma to neutron) is 1.17. The relative standard deviation in the assay ratio is 28%. The relative uncertainty in the neutron holdup assay is estimated to be approximately 10%. Thus, the observed relative standard deviation is attributed to the uncertainty in the gamma-ray holdup assay.

The major contributor to the uncertainty in the gamma-ray holdup assay is the uncertainty (~25 %) in the correction factors for equipment attenuation. The assumption that the holdup is deposited at the center of the process equipment could also result in a systematic component (a positive bias, in this case, resulting from an overestimate of equipment attenuation effects) in this uncertainty. Ignoring self-attenuation effects is an obvious source of a systematic effect (negative bias in this case), but the measured deposit thicknesses indicate that this is small relative to the uncertainties in the equipment attenuation. The calibration uncertainty that results from variations in the isotopic distributions (varying interference effects) is estimated to be approximately 10%. Measurement precision is negligible in this discussion. Other uncertainties (in room backgrounds, detector positioning, and nonuniform deposit geometries) are also expected to be relatively small. The use of high-resolution gamma-ray spectroscopy would eliminate the 10% calibration uncertainty. However, only an improved knowledge of the equipment attenuation effects, which would certainly be achieved with gamma-ray holdup measurements of ducts or piping, would lead to a significant improvement in the assay uncertainty.

TABLE II. Original Analysis of Glove-Box Data

Glove Box	Average R_a (s^{-1})	1σ (%)	^{239}Pu Wt Fraction	CF Steel (400 keV)	Assay Ratio (Gamma/Neutron)
UCP01	17.5	3	0.63	1.5	0.73
UCP03-w	7.9	7	0.63	1.5	
UCP03-a	23.6	3	0.63	1.2	1.30
UCP04	34.3	1	0.61	1.2	
UCP04-2	161.4	1	0.61	3.0	0.90
UFP01	24.1	2	0.63	1.2	1.24
UFP02-w	58.3	1	0.63	1.5	
UFP02-a	16.5	4	0.63	1.5	1.57
UFP03	21.7	3	0.62	1.5	1.46
UFP05-w	7.6	16	0.63	2.0	
UFP05-a	30.6	3	0.63	2.0	1.56
UFP12-w	64.8	2	0.62	2.0	
UFP12-a	43.4	2	0.62	1.2	1.05
UPF15	3.9	13	0.63	1.2	0.74
w—Glove-box section adjacent to wall				Av =	1.17
a—Glove-box section adjacent to aisle				1σ (%) =	28

These low-resolution gamma-ray holdup measurements of high-burnup plutonium were successful because they agreed within expectations with the neutron holdup assays. However, this success is largely the result of the range of standards available for the calibration. The major drawback to the appmach is the dependence of the calibration on the evaluation of spectra produced by process samples because the signature for ^{239}Pu assay contains a variable ^{241}Am interference. A new analysis method is required that eliminates the interference from the assay signature and requires only one standard for calibration.

III. A NEW LOW-RESOLUTION PLUTONIUM SPECTRAL ANALYSIS METHOD

A. Introduction

The assay ROI for the low-resolution gamma-ray assay of ^{239}Pu is dominated by activity from ^{239}Pu, but also contains significant activity from ^{241}Am as the sample burnup and age increase. The new analysis procedure corrects the measured activity in the assay region for the ^{241}Am interference on a spectrum by spectrum basis. The procedure combines measured activities from energy regions below the assay ROI (ROIa) with theoretical activity ratios to obtain a unique ^{241}Am correction for each spectrum.

Two new energy ROIs are defined below ROIa, at 300 to 375 keV and at 181 to 235 keV. These two new ROIs are highlighted in the four spectra in Fig. 1. The continuum background for ROI3 (300 to 375 keV) is the same as that used for ROIa. Continuum background for ROI2 (181 to 235 keV) was obtained from two narrow (6- to 7-keV-wide) ROIs, not highlighted in Fig. 1, set on either side of ROI2.

A set of seven, physically identical reference standards,[4] P1 through P7, were used to develop and test the theoretical basis of the new analysis procedure. These and an additional set of four physically identical standards,[5] C8 through C11, were used to evaluate the new analysis procedure. Table I gives the isotopic compositions and plutonium masses for the standards. In both sets, burnup (^{241}Pu content) increases with standard number.

B. Derivation of Theoretical Activity Ratios

Theoretical gamma-ray isotopic activities were calculated for ROI2, ROI3, and ROIa (181 to 235 keV, 300 to 375 keV, and 375 to 450 keV, respectively) for the standards, P1 through P7. The activity (in s^{-1}) for a gamma ray of a given isotope is given by

$$I = \frac{N \cdot \lambda}{\alpha} \cdot B \cdot m \ , \qquad (1)$$

where

- N = 6.023×10^{23} atoms/mole,
- λ = isotope decay constant (s^{-1}),
- α = isotope atomic weight (g/mole),
- B = gamma-ray branching ratio (gamma rays/disintegration), and
- m = isotope mass (g).

The isotopes that contribute to the activities in the three energy regions are ^{239}Pu, ^{241}Am, and ^{237}U. The ^{237}U is in secular equilibrium with ^{241}Pu for the standards as well as for the process material holdup. The masses of ^{239}Pu, ^{241}Pu, and ^{241}Am in the standards can be computed from Table I as the product of plutonium mass and the respective

isotope weight fractions. The ^{237}U mass is computed from the ^{241}Pu mass as follows:

$$m_{237} = \frac{\lambda_{241} \cdot \alpha_{237}}{\lambda_{237} \cdot \alpha_{241}} \cdot m_{241} \ , \qquad (2)$$

where the subscripts 237 and 241 refer to the ^{237}U and ^{241}Pu isotopes, respectively, and the ^{241}Pu decay constant is the partial decay constant for the alpha decay branch.

The total theoretical activity in a region was computed for each isotope by summing the activities of all gamma rays within the region separately for each isotope. The summed activities are plotted vs standard number for the three isotopes (along with the total for all three) in Fig. 2(a), (b), and (c) for ROI2, ROI3, and ROIa, respectively. Assuming that variations in detection efficiency within a region and the partial overlap of some of the low-resolution (finite-width) peaks with the energy region can be ignored to first order, these plots show the theoretical relative contributions of the isotopes to the low-resolution gamma-ray spectra of the standards for the three energy regions.

Figure 2(c) shows the increase in ^{241}Am interference with increasing burnup for the assay ROI. This effect becomes larger still as the samples age. Figure 2(b) shows that ROI3 includes significant components of all three isotopes at most burnups. However, ROI2 is dominated by ^{237}U activity at all burnups, as shown in Fig. 2(a). The new procedure uses the measured activity in ROI2 to determine and remove the ^{241}Pu–^{237}U activity from ROI3. Only ^{241}Am and ^{239}Pu activities remain in ROI3 and ROIa.

The theoretical ratio of ^{241}Pu–^{237}U gamma-ray activities between ROI2 and ROI3 for the NaI(Tl) holdup detector is

$$r = \frac{\sum_i B_i \cdot (1 - T_{336}) \cdot T_{336}^{-1}}{\sum_j B_j \cdot (1 - T_{208}) \cdot T_{208}^{-1}} \ , \qquad (3)$$

where

$\sum_i B_i$ = ^{241}Pu–^{237}U branching ratio sum from 300 to 375 keV,

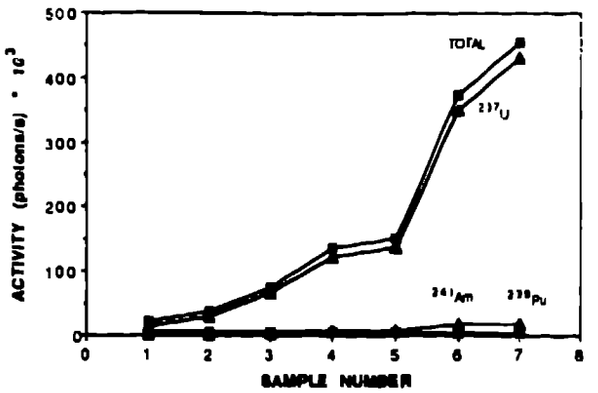
$\sum_j B_j$ = ^{241}Pu–^{237}U branching ratio sum from 181 to 235 keV,

T_n = n-keV-gamma-ray transmission through 5 cm of NaI, and

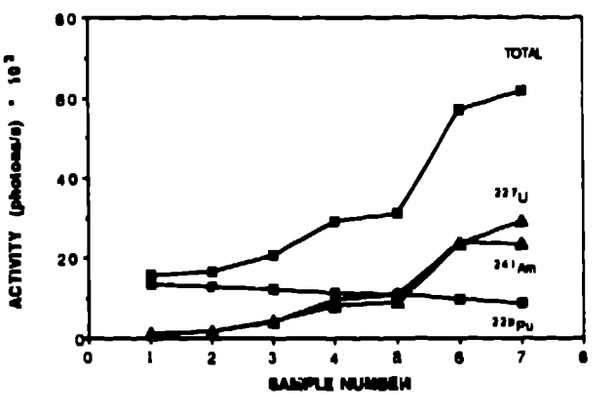
T_n' = n-keV-gamma-ray transmission through 1.6 cm of cadmium.

The gamma-ray transmission expression is

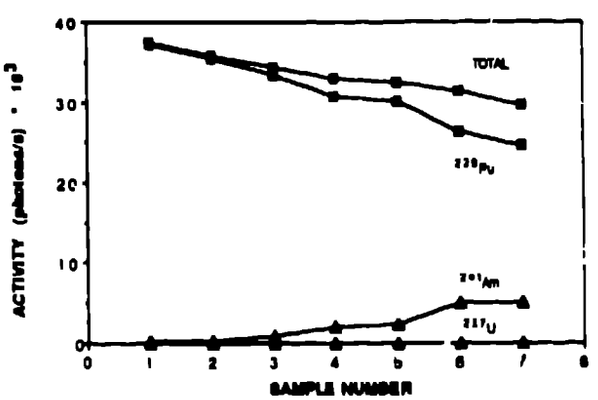
$$T = e^{\mu x} \ , \qquad (4)$$



(a)



(b)



(c)

Fig. 2(a)–(c). Theoretical isotopic components of the ROI gamma-ray activities for the standards P1–P7 plus the total ROI gamma-ray activity, plotted vs standard number, for (a) ROI2, (b) ROI3, and (c) the assay ROI, ROIa.

where ρ and t are the density (g/cm^2) and thickness (cm), respectively, of the material and μ is the mass attenuation coefficient (cm^2/g) of the material at the energy of the gamma ray. The energies, n, are the mean energies (corresponding to the approximate centroids of the distributions of net counts) within the ROIs.

The theoretical ratios, analogous to Eq. (3), of ^{241}Am and ^{239}Pu gamma-ray activities between ROI3 and ROIa for the NaI(Tl) holdup detector are

$$y = \frac{\sum_j B_j^{Am} \cdot (1 - T_{336}) \cdot T_{336}^{-1}}{\sum_j B_j^{Am} \cdot (1 - T_{410}) \cdot T_{410}^{-1}} \qquad (5)$$

and

$$z = \frac{\sum_j B_j^{239} \cdot (1 - T_{336}) \cdot T_{336}^{-1}}{\sum_j B_j^{239} \cdot (1 - T_{410}) \cdot T_{410}^{-1}} , \qquad (6)$$

respectively, where

$\sum_j B_j^{Am}$ = ^{241}Am branching ratio sum from 300 to 375 keV,

$\sum_j B_j^{Am}$ = ^{241}Am branching ratio sum from 375 to 450 keV,

$\sum_j B_j^{239}$ = ^{239}Pu branching ratio sum from 300 to 375 keV, and

$\sum_j B_j^{239}$ = ^{239}Pu branching ratio sum from 375 to 450 keV.

The coefficients x, y, and z are used to derive the ^{241}Pu-^{237}U, ^{241}Am and ^{239}Pu components of the net counts in ROI3 and ROIa of each spectrum. The ^{239}Pu component of ROIa is the ^{239}Pu assay signal, independent of ^{241}Am interference. The ratio of the ^{241}Pu-^{237}U component to the ^{239}Pu component is a measure of the burnup attribute. The ratio of the ^{241}Am component to the ^{241}Pu-^{237}U component is a measure of the age attribute.

C. Evaluation of New Analysis Method with Standards

Gamma-ray spectra were obtained with the NaI(Tl) holdup detector for the standards, P1– P7 and C8–C11, described in Table I. (Figure 1 shows these spectra for P1 and P4–P6.) The isotopic components of ROI3 and ROIa were evaluated for each of these spectra.

As a first step in this procedure, the measured net count rates in ROI2, ROI3, and ROIa are multiplied by correction factors for source self-attenuation and attenuation by the source cladding. The attenuation corrections are performed at the ROI mean energies, 208, 336, and 410 keV, respectively. The self-attenuation correction factor for the 11 pellet-shaped standards, viewed end-on at a large distance compared to the pellet dimensions, is

$$CF_{self} = \ln T_n^s / (T_n^s - 1) , \qquad (7)$$

where

T_n^s = n-keV-gamma-ray transmission through the source.

The cladding attenuation correction factor is

$$CF_{clad} = 1/T_n^c , \qquad (8)$$

where

T_n^c = n-keV-gamma-ray transmission through the cladding.

Equation (4) gives the form of T_n^s and T_n^c.

In the second step, the ^{241}Pu–^{237}U component is subtracted from the ROI3 net count rate to give

$$R_3' = R_3 - x \cdot R_2 , \qquad (9)$$

where

R_k = measured, attenuation-corrected ROIk net count rate

and

R_k' = adjusted ROIk net count rate.

Two more equations can now be written:

$$R_3' = R_3^{Am} + R_3^{239} \qquad (10)$$

and

$$R_a = R_a^{Am} + R_a^{239} , \qquad (11)$$

where the superscripts, Am and 239, label the ^{241}Am and ^{239}Pu components of the net count rates in ROI3 and ROIa. The isotopic components represent four unknowns in the two Eqs., (10) and (11).

The third and final step is to reduce the number of unknowns in Eqs. (10) and (11) to two and to subsequently solve the equations to give the ^{241}Am and ^{239}Pu components of the net count rates in ROI3 and ROIa. Rewriting the Eq. (10) unknown components in terms of the Eq. (11) unknown components gives

$$R_3^{Am} = y \cdot R_a^{Am} \qquad (12)$$

and

$$R_3^{239} = z \cdot R_a^{239} , \qquad (13)$$

where y and z are defined in Eqs. (5) and (6), respectively. Substituting Eqs. (12) and (13) into Eq. (10) and solving for the two independent ROIa components gives

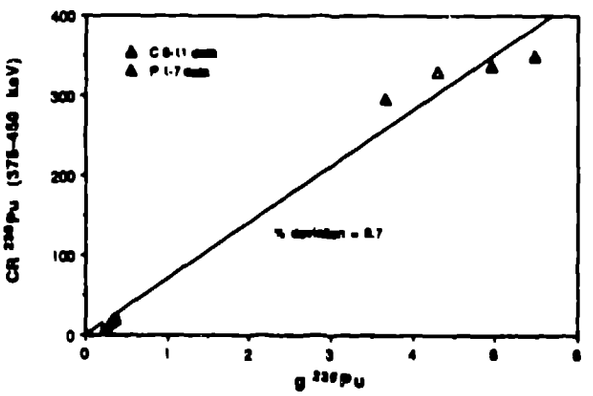
$$R_a^{Am} = \left(R_3' - z \cdot R_a\right)/(y - z) \qquad (14)$$

and

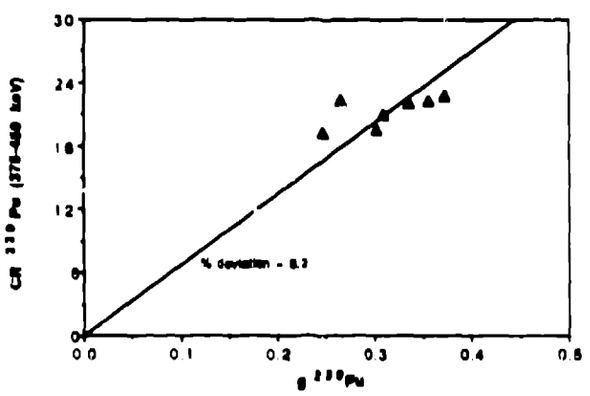
$$R_a^{239} = R_a - R_a^{Am} \ , \qquad (15)$$

where R_a^{Am} is defined by Eq. (14).

Equations (14) and (15) give the ^{241}Am and ^{239}Pu count rate components of the attenuation-corrected net count rate in ROIa, the assay region. These are proportional to the ^{241}Am and ^{239}Pu masses, respectively. The original analysis used the count rate, R_a, including the ^{241}Am component, as the signal for the ^{239}Pu assay. Figures 3(a), (b) and 4(a), (b) are plots of the ^{239}Pu assay signal for the original and new

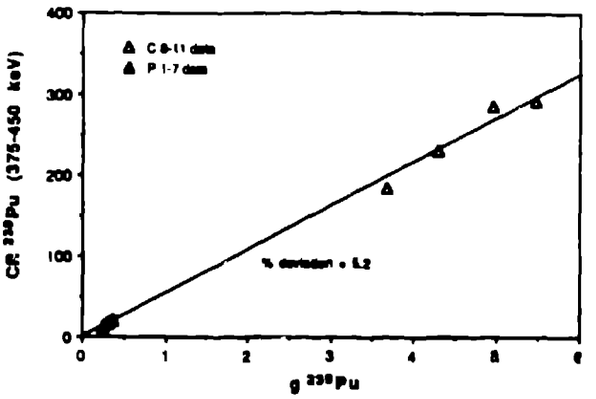


(a)

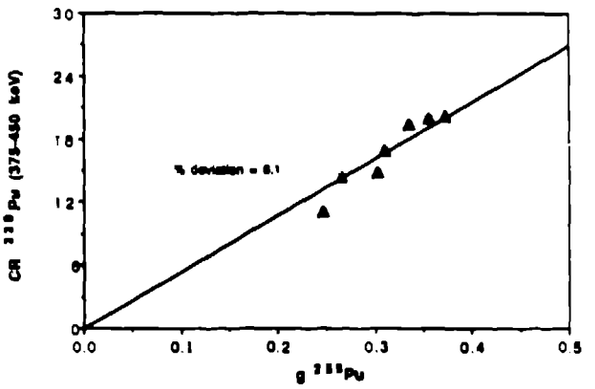


(b)

Fig. 3(a), (b). Net count rate in the assay ROI, ROIa, determined by the single-ROI method vs ^{239}Pu mass for the standards (a) P1–P7 and C8–C11 and for (b) P1–P7 only. The straight line is a linear fit to the data. The relative average absolute deviation of the fit is shown.



(a)



(b)

Fig. 4(a), (b). Net ^{239}Pu count rate in the assay ROI, ROIa, calculated by the three-ROI method vs ^{239}Pu mass for the standards (a) P1–P7 and C8–C11 and for (b) P1–P7 only. The straight line is a linear fit to the data. The relative average absolute deviation of the fit is shown.

analysis methods, respectively, vs the reference ^{239}Pu mass for all of the standards. The departures from linearity in the original analysis are most evident for the lowest-^{239}Pu-mass (highest burnup) standards in each set.

The quantity R_2, the attenuation-corrected net count rate in ROI2, is proportional to the ^{241}Pu mass. Therefore, the ratio of R_2 to Eq. (15), which is proportional to the ^{241}Pu-to-^{239}Pu isotope ratio, is a signal for the burnup attribute. The ratio of Eq. (14) to R_2, which is proportional to the ^{241}Am-to-^{241}Pu isotope ratio, is a signal for the age attribute. Figure 5 is a plot of the experimental ^{241}Pu-to-^{239}Pu isotope ratio vs the reference value for the 11 standards. The average, absolute deviation of the linear fit from the data is 7%, including the two low-burnup (P1 and C8) standards and 5% excluding them. These experimental data for the lowest burnup standards are positively biased because of the 203-keV ^{239}Pu interference (of ~25%) in the ROI2 net rate. This effect is much smaller at higher

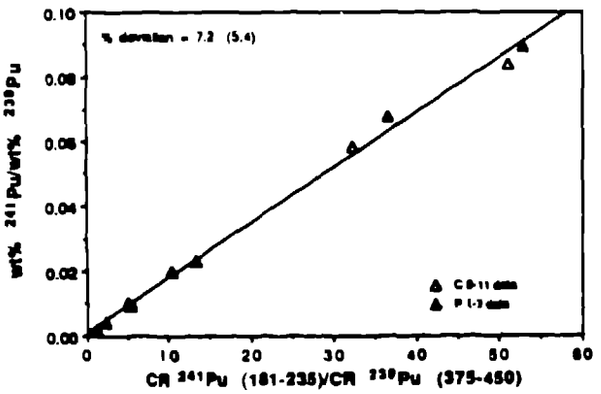


Fig. 5. The reference values for the ^{241}Pu-to-^{239}Pu mass ratios are plotted vs the ^{241}Pu-to-^{239}Pu count rate ratios determined by the three-ROI method for the standards P1–P7 and C8–C11. The straight line is a linear fit to the data. The relative average absolute deviation of the fit is shown, with and without (in parentheses) the lowest burnup (P1 and C8) results included.

burnups. A similar comparison of the experimental ^{241}Am-to-^{241}Pu ratios with reference values gives a large (45%) average absolute deviation, primarily because the ^{241}Am activity is derived from ROIs dominated by other activities.

D. Summary of New Analysis Procedure

Only a single point-source standard is required to obtain the calibration of the generalized-geometry holdup assay of ^{239}Pu, independent of ^{241}Am interference. The point-source standard with ^{239}Pu mass, m_0 (g), is measured at the distance, r_0 (cm), on the detector axis. The value of $R_{a,0}^{239}$ (R_a^{239} for the standard) is obtained by solving Eqs. (9), (14), and (15) using the theoretically derived x, y, and z values for the detector/spectral analysis system and the measured, attenuation-corrected net counts in ROI2, ROI3, and ROIa. The absolute calibration response per unit mass of ^{239}Pu at r_0,

$$k = R_{a,0}^{239}/m_0 \qquad (s^{-1}\,g^{-1}) , \qquad (16)$$

is used to obtain the specific ^{239}Pu holdup masses in the respective point, line, or area deposit geometries, measured at the distance, r (cm), as follows:

$$m_{point} = (R_a^{239}/k)\,(r/r_0)^2 \qquad (g\ ^{239}Pu) , \qquad (17)$$

$$m_{line} = (R_a^{239}/L \cdot k)(r/r_0) \qquad (g\ ^{239}Pu/cm) , \qquad (18)$$

and

$$m_{area} = (R_a^{239}/A \cdot k) \qquad (g\ ^{239}Pu/cm^2) . \qquad (19)$$

The detector-specific geometric factors, L (cm) and A (cm^2), for line- and area-source measurements, respectively, are defined elsewhere.[1,2,9] Equations (16) through (19), including all terms, are the same equations used to calibrate the assay and quantify the results for the original PFPF holdup measurements except that $R_{a,0}$ and R_a from Eq. (11) were used for the calibration and assay signals instead of $R_{a,0}^{239}$ and R_a^{239}, respectively.

IV. NEW ANALYSIS APPLIED TO HOLDUP MEASUREMENT SPECTRA

The stored low-resolution gamma-ray spectra from the generalized-geometry holdup calibration and the glove-box holdup measurements were re-analyzed using the new analysis procedure. The results of the re-analysis are given in Table III. These include the average ^{241}Am-corrected net count rate in ROIa (R_a^{239}, s^{-1}), its relative uncertainty, and the ratio of gamma-ray to neutron assays for each glove box.

The notable decreases in the gamma-ray to neutron assay ratios for UCP03, UCP04, UFP01, UFP05, and UFP12 in Table III compared to the results in Table II are likely to be the effects of differences in the americium weight fractions of the holdup deposits. For glove boxes in which the americium fraction of the holdup deposits is greatest, the original gamma-ray assay based on the single ROI analysis will be positively biased relative to assays for glove boxes with lower americium fractions in the holdup deposits. The assay based on the new analysis will be unbiased as a result of this difference. Another possible cause of decreased gamma-ray to neutron assay ratios is a significant excess of ^{237}Np in the holdup deposits for these glove boxes relative to the others. Because the ^{237}Np activity is concentrated in the ROI3 energy range, its presence at levels above 1000 parts per million (by weight, relative to plutonium) results in a significant oversubtraction of the ^{241}Am activity from the measured ROIa activity.

The measured net count rates in all three ROIs, including the two new ROIs (ROI2 and ROI3), were corrected for equipment attenuation effects in the new analysis procedure. This correction, which was performed in the original analysis only for the ROIa net count rate, is significantly larger at the lower energies, especially at 208 keV. The steel attenuation correction factors at the mean ROI2 and ROI3 energies are given in Table III. Because the average steel attenuating thickness is only an estimate for automated equipment of this type, and because some of the estimated thicknesses are quite large (16 mm for the calciner, UCP04-2), additional systematic effects resulting from larger uncertainties in the equipment attenuation correction factors are expected. However, agreement with the neutron holdup assays remains reasonable (within 10% with a 35% relative standard deviation), as shown by the average gamma-ray-to neutron assay ratio in Table III.

TABLE III. Reanalysis of Glove-Box Data

Glove Box	Average R_q^{239} (s^{-1})	1σ (%)	CF Steel (208 keV)	CF Steel (336 keV)	Assay Ratio (Gamma/Neutron)
UCP01	8.8	7	1.9	1.6	0.70
UCP03-w	5.1	11	1.9	1.6	
UCP03-a	9.0	6	1.4	1.3	1.06
UCP04	12.5	2	1.4	1.3	
UCP04-2	32.1	4	4.7	3.2	0.52
UFP01	9.2	5	1.4	1.3	0.89
UFP02-w UFP02-a	Spectral data were not available for reanalysis				
UFP03	12.1	6	1.9	1.6	1.53
UFP05-w	3.0	30	2.8	2.2	
UFP05-a	11.8	8	2.8	2.2	1.14
UFP12-w	22.4	4	2.8	2.2	
UFP12-a	19.1	3	1.4	1.3	0.72
UPF15	2.0	22	1.4	1.3	0.73
w—Glove-box section adjacent to wall a—Glove-box section adjacent to aisle				Av = 0.91 1σ (%) = 35	

V. CONCLUSIONS AND RECOMMENDATIONS

The quantitative assay of high-burnup plutonium holdup by low-resolution gamma-ray spectrometry with a single assay ROI has been achieved by calibrating the assay with four plutonium standards representing a range of fuel burnup and ^{241}Am content. A new procedure that requires only one plutonium calibration standard and uses three ROIs has been developed and tested with standards and with spectra obtained from high burnup-plutonium holdup deposits. The new procedure uses calculated theoretical isotope activity ratios for the three energy ROIs. The total ^{239}Pu mass, as well as the burnup attribute of the high-burnup material, are obtained with the new procedure.

Improvements to the calculated theoretical isotope activity ratios include folding finite spectral widths into the branching ratio summations and incorporating the detection efficiency effects on a (gamma-ray) line-by-line basis, rather than applying gross factors to the energy-region branching-ratio summation. The addition of higher-americium standards (requiring interference corrections in excess of 30%) to the test materials for the new procedures would be useful for sensitive evaluation of these improvements. An iterative correction for the ^{239}Pu 203-keV interference with the ^{241}Pu 208-keV activity would improve the accuracy of experimentally determined isotope ratios for low-burnup materials.

The application of these gamma-ray holdup assay procedures for process materials consisting of high-burnup plutonium is best suited to equipment such as pipes, ducts, and tanks, for which thicknesses of attenuating equipment are relatively well known and uniform. In these cases, the small, collimated, shielded gamma-ray detector is also a better choice than the large neutron slab detectors because measurements of this type of equipment often involve access difficulties, high equipment densities and low signals in high-background areas. Testing of the new scintillator-photodiode detectors in these applications is recommended for increased portability and simplicity.

The new procedure has been applied to gamma-ray spectra from reference materials and holdup deposits with ^{241}Am components near 5% by weight. The subtraction of the ^{241}Am interference results in losses in assay precision. For this reason, the procedure is not recommended for assay of materials for which the ^{241}Am component exceeds 10% by weight. Because of the potential for bias, the procedure is not recommended for assay of materials in which the ^{237}Np weight fraction exceeds 1000 parts per million. Assay of high-burnup plutonium holdup deposits for which either of these limits is exceeded can be accomplished by using the 208-keV ^{241}Pu–^{237}U gamma-ray peak as the assay signal. The two shortcomings of this approach are the larger uncertainty in the ^{241}Pu weight fraction (compared to that for ^{239}Pu) of the holdup deposits and the significantly larger effects of uncertainties in equipment attenuation correction factors. For holdup measurements of process piping, ducts, or tanks, the latter problem is minimal, even for the lower energy assay.

The new procedure is simple to implement, despite requirements for analyzing additional energy ROIs. The sensitivity to interference effects for both (new and original) procedures is minimized by a conservative choice of limits for the assay ROI. However, this choice also increases the sensitivity of the assay to the effects of gain drift. Therefore, these low-resolution methods applied to high-burnup plutonium derive a particular benefit from automated gain drift compensation. Automation is particularly desirable for

the new procedure in the real-time reduction and analysis of multiple spectral regions of interest.

The extension of the low-resolution gamma-ray spectral analysis for determination of high-burnup plutonium quantities and attributes to multiple energy regions of interest generalizes the calibration for plutonium of any burnup or age. A further extension of this analysis by application of pattern-recognition techniques[10] to the low-resolution spectra may offer more precise results using the same gamma-ray spectra. These efforts are in progress.

VI. ACKNOWLEDGMENTS

Two of the authors (T. R. Wenz and P. A. Russo) thank Jack L. Parker and Thomas E. Sampson for informative discussions and suggestions on the procedures. The assistance of Ted Dye and Jo Ann Painter with hardware and software requirements (respectively) for this project is gratefully acknowledged.

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