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FEASIBILITY STUDY OF PLUTONIUM ISOTOPIC ANALYSIS OF RESIN BEADS BY NONDESTRUCTIVE GAMMA-RAY SPECTROSCOPY

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Abstract

We have initiated a feasibility study on the use of nondestructive low-energy gamma-ray spectroscopy for plutonium isotopic analysis on resin beads. Seven resin bead samples were measured, with each sample containing an average of 9 μg of plutonium; the isotopic compositions of the samples varied over a wide range. The gamma-ray spectroscopy results, obtained from 4-h counting-time measurements, were compared with mass spectrometry results. The average ratios of gamma-ray spectroscopy to mass spectrometry were 1.014 ± 0.025 for $^{238}Pu/^{239}Pu$, 0.996 ± 0.018 for $^{240}Pu/^{239}Pu$, and 0.980 ± 0.038 for $^{241}Pu/^{239}Pu$. The rapid, automated, and accurate nondestructive isotopic analysis of resin beads may be very useful to process technicians and International Atomic Energy Agency inspectors.

1. Introduction

We recently developed a nondestructive gamma-ray technique[1] to accurately determine plutonium isotopic ratios in a timely manner for plutonium having a wide range of masses, isotopic contents, and chemical forms. This technique, based on high-intensity, low-energy (36-68 keV) gamma rays, has demonstrated its sensitivity in measuring very small mass samples. For example, within a 10-min counting time, the measured precision for a 15-mg reactor-grade plutonium sample is 1.5% for $^{238}Pu/^{239}Pu$, 0.5% for $^{240}Pu/^{239}Pu$, and 6.1% for $^{241}Pu/^{239}Pu$. Following a suggestion offered by Higinbotham[2], we recently applied this technique to initiate a feasibility study of plutonium isotopic analysis on anion resin beads. The resin bead–mass spectrometric technique involves absorption of plutonium into resin beads and subsequent mass spectrometric analysis was developed at Oak Ridge National Laboratory[3]. This paper reports the results of the first measurements obtained by a nondestructive gamma-ray technique on resin beads.

2. Measurement Method

Details of the measurement technique are described in Ref. 1. Briefly, the isotopic ratios of $^{238}Pu/^{239}Pu$, $^{240}Pu/^{239}Pu$, and $^{241}Pu/^{239}Pu$ are determined by analyzing the 43.48 keV/51.63 keV, 45.23 keV/51.63 keV, and 148.6 keV/129.3 keV gamma-ray ratios, respectively. All the gamma-ray peak areas are calculated using a channel summation method with a linear background subtraction. Minor interferences in the full-energy peaks are taken into account. The gamma-ray relative efficiencies are determined using the measured peak areas and the known specific activities of ^{239}Pu gamma rays at energies of 38.66, 51.63, 68.72, 129.3, 144.2, 171.3, 195.7, and 203.5 keV.

The measurement system consists of a high-resolution hyperpure germanium planar detector and associated electronics, a Canberra Series 90 multichannel analyzer (MCA) with an 8-k channel analog-to-digital converter (ADC), and a Digital Equipment Corporation (DEC) LSI-11/23 microcomputer and peripherals. The MCA is controlled by the LSI-11/23 microcomputer, which has 128-k 16-bit words of memory and is a processor for data acquisition and analysis. Dual DEC RL01 cartridge disks provide for storage and transfer of the program and data. The control of assay input and output is accomplished through a DEC VT100 video terminal. The hyperpure germanium planar detector has dimensions of 1000 mm^2 x 13 mm and a resolution (full width at half maximum) of 560 eV at 122 keV. An automatic data-acquisition-and-analysis program is written in FORTRAN under DEC's RT-11 V-4.0 operating system. For setting up routine assays, the operator/computer dialogue is kept as simple as possible.

3. Results and Discussion

Two sets of resin bead samples with completed mass spectrometer analyses were provided by Joel A. Carter, Oak Ridge National Laboratory, and are described in Table I. Set I (samples Mix 1 to Mix 4) was prepared on November 15, 1983, and Set II (samples PA966 to PA968) was prepared on May 14, 1984. Each sample consists of ~10 000 beads loaded with an average of 9 μg of plutonium contained in a small plastic vial. Isotopic compositions varied from 0.003 to 1.41% for ^{238}Pu, from 57.53 to 97.47% for ^{239}Pu, from 2.49 to 36.23% for ^{240}Pu, and from 0.027 to 8.3% for ^{241}Pu.

Figure 1 shows the low-energy (38-to 60-keV) gamma-ray spectra from three resin bead samples. The spectra were taken approximately one month after the resin bead samples were prepared. The 59.54-keV gamma rays from ^{237}U and ^{241}Am (decay of ^{241}Pu) are prominent in the spectra as a function of the ^{241}Pu content in the samples, but

TABLE I

PLUTONIUM ISOTOPIC COMPOSITION (in at.%)

Resin Bead Sample	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
Mix 1	0.003	97.47	2.49	0.039	0.003
Mix 2	0.017	81.04	18.90	0.032	0.006
Mix 3	0.024	72.55	27.39	0.029	0.008
Mix 4	0.033	63.70	36.23	0.027	0.009
PA966	0.055	87.02	11.70	1.022	0.206
PA967	0.276	77.27	18.71	2.54	1.21
PA968	1.41	57.53	27.37	8.30	5.39

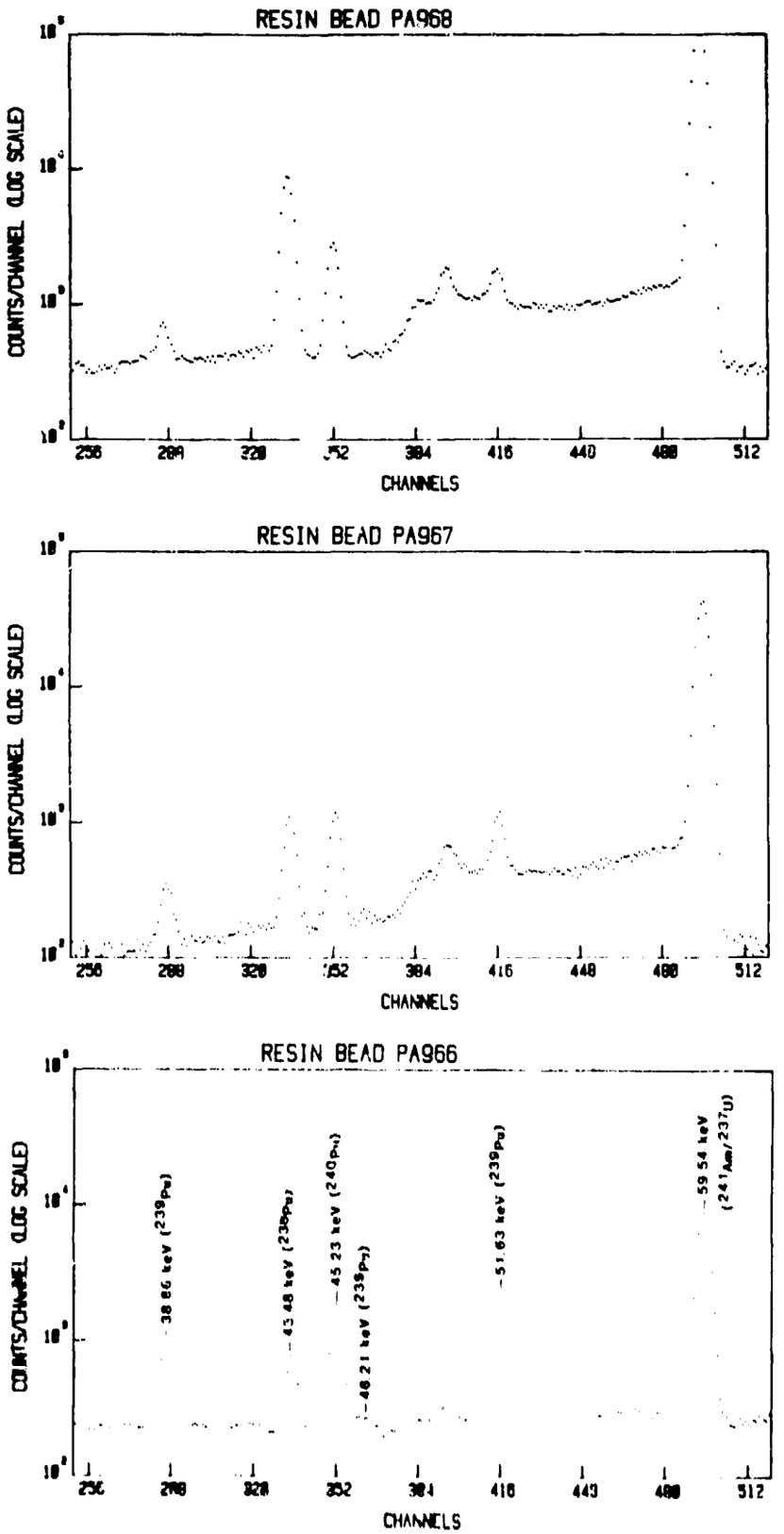

Fig. 1. Low-energy (38- to 60-keV) gamma-ray spectra from resin bead samples PA966, PA967, and PA968. These spectra were taken approximately one month after the samples were prepared.

have only a minor effect on the analysis of low-energy gamma rays at 43.48, 45.23 and 51.63 keV.

Table II compares the preliminary gamma-ray spectroscopy results for a 4-h count time with mass spectrometry results. The uncertainties represent the estimated precision (1σ) of gamma-ray ray spectroscopy as calculated from counting statistics, including uncertainties from relative efficiencies. The average ratios of gamma spectroscopy to mass spectrometry are 1.014 for $^{238}Pu/^{239}Pu$, 0.996 for $^{240}Pu/^{239}Pu$, and 0.980 for $^{241}Pu/^{239}Pu$. Because the $^{241}Pu/^{239}Pu$ ratios for samples Set I (Mix 1 to Mix 4) are so small ($\sim 4.0 \times 10^{-4}$), no attempt has been made to analyze them. These results show negligible bias when compared with mass spectrometry results. The 2.5%, 1.8%, and 3.8% standard deviations of

TABLE II

COMPARISON OF ISOTOPIC RATIOS BY GAMMA-RAY SPECTROSCOPY[a] WITH MASS SPECTROMETRY

Isotopic Ratio	Resin Bead Sample	Mass Spectrometry	Gamma-Ray Spectroscopy	Gamma-Ray Spectroscopy / Mass Spectrometry
$^{238}Pu/^{239}Pu$				
	Mix 1	3.05×10^{-5}	$3.11 \times 10^{-5} \pm 70\%$[b]	1.020
	Mix 2	2.08×10^{-4}	$2.06 \times 10^{-4} \pm 16.3\%$	0.990
	Mix 3	3.28×10^{-4}	$3.37 \times 10^{-4} \pm 10.9\%$	1.027
	Mix 4	5.13×10^{-4}	$5.04 \times 10^{-4} \pm 7.2\%$	0.983
	PA966	6.29×10^{-4}	$6.24 \times 10^{-4} \pm 6.0\%$	0.992
	PA967	3.56×10^{-3}	$3.70 \times 10^{-3} \pm 2.2\%$	1.039
	PA968	2.44×10^{-2}	$2.55 \times 10^{-2} \pm 5.2\%$	1.045
	Average			1.014
	Standard Deviation			±0.025
$^{240}Pu/^{239}Pu$				
	Mix 1	0.0257	0.0256 ± 6.6%	0.996
	Mix 2	0.234	0.230 ± 2.1%	0.983
	Mix 3	0.379	0.387 ± 2.2%	1.021
	Mix 4	0.571	0.563 ± 2.0%	0.986
	PA966	0.135	0.138 ± 2.3%	1.022
	PA967	0.243	0.238 ± 2.0%	0.979
	PA968	0.478	0.471 ± 4.6%	0.985
	Average			0.996
	Standard Deviation			±0.018
$^{241}Pu/^{239}Pu$[c]				
	PA966	0.0118	0.0113 ± 18.3%	0.956
	PA967	0.033	0.0338 ± 10.5%	1.024
	PA968	0.145	0.139 ± 21.9%	0.959
	Average			0.980
	Standard Deviation			±0.038

[a]Counting time = 4 h.
[b]Uncertainties represent the estimated precision (1σ) as calculated from counting statistics, including uncertainties from relative efficiencies.
[c]Because the $^{241}Pu/^{239}Pu$ ratios for samples Mix 1 to Mix 4 are so small ($\sim 4.0 \times 10^{-4}$), no attempt has been made to analyze them.

$^{238}Pu/^{239}Pu$, $^{240}Pu/^{239}Pu$, and $^{241}Pu/^{239}Pu$, respectively, indicate the overall precision of measurements by low-energy gamma-ray spectroscopy in these wide ranges of plutonium isotopic distribution (within a 4-h count time). Obviously, the precision obtained from the gamma-ray technique is affected by counting statistics of the full-energy peaks; the counting statistics are a function of sample mass, isotopic distribution, assay geometry, count time, sample age, and so forth.

Table III lists the measured precision (as determined from 15 repeated measurements) of seven resin bead samples measured by gamma-ray spectroscopy in 4-h count times. These results demonstrate that this technique can be applied to various types of materials in reprocessing plants. For example, sample PA966 is typical of FFTF plutonium feed, and samples PA967 and PA968 are typical of plutonium from spent fuel in LMFBR and LWR fuel cycles. The measured precisions in 4-h count time for these samples are from 2.4 to 4% for $^{240}Pu/^{239}Pu$ and from 3.2 to 6.3% for $^{238}Pu/^{239}Pu$. The precision can be improved by placing the samples closer to the detector and by measuring the samples as soon as they are prepared at the plant (before the 59.54-keV peak starts to grow).

Verification of input plutonium and assay of samples from intermediate processes are very important for near-real-time accounting at reprocessing plants. At present, International Atomic Energy Agency (IAEA) inspectors must spend many hours to observe the preparation of samples to be sent to Vienna for analysis. Higinbotham[2] has suggested that simple, rapid, at-plant, attribute analyses to a few percent be performed, so that only a few accurate variables assays need be done at the SAL in Vienna. The attribute sample analyses should flag major problems in time to resolve them with little effort. It is understood that typically several thousand beads containing several micrograms of plutonium are loaded at a time, and only a few beads would be shipped to Vienna. The rest could be used for gamma-ray analysis for easy prompt verification. Inspectors could then spend less time observing chemical operations to detect altering or substitution of samples.

In summary, nondestructive gamma-ray spectroscopy has been used for the first time to measure resin beads. The rapid and accurate nondestructive isotopic analysis of resin beads may provide useful information on burnup, accountability, process monitoring, and so forth. Furthermore, the simple operation of automated nondestructive gamma-ray instruments may be of interest to process technicians (for routine measurements in reprocessing facilities) and IAEA inspectors. Further studies of spiked samples to determine plutonium and uranium concentrations are under way.

TABLE III

MEASURED PRECISION (1σ, %) OF PLUTONIUM ISOTOPIC RATIOS IN 4-h ASSAY TIME

Resin Bead Sample	$\frac{^{238}Pu}{^{239}Pu}$	$\frac{^{240}Pu}{^{239}Pu}$	$\frac{^{241}Pu}{^{239}Pu}$
Mix 1	30.	6.6	
Mix 2	15.	1.4	
Mix 3	13.	2.5	
Mix 4	8.1	2.5	
PA966	6.3	2.5	19.9
PA967	3.2	2.4	12.8
PA968	4.2	4.0	15.7

4. Acknowledgments

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5. References

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