PLUTONIUM ISOTOPIC MEASUREMENTS BY GAMMA-RAY SPECTROSCOPY

Francis X. Haas and John F. Lemming

Monsanto Research Corporation
Mound Laboratory*
Miamisburg, Ohio

Presented at the
ERDA Symposium on X- and Gamma-Ray
Sources and Applications
Ann Arbor, Michigan
May 19-21, 1976

*Mound Laboratory is operated by Monsanto Research Corporation for the U. S. Energy Research and Development Administration under Contract No. E-33-1-GEN-53.

PLUTONIUM ISOTOPIC MEASUREMENTS BY GAMMA-RAY SPECTROSCOPY

F. X. Haas and J. F. Lemming
Monsanto Research Corporation, Mound Laboratory*
Miamisburg, OH 45342

The nondestructive assay of plutonium is important as a safeguard tool in accounting for strategic nuclear material. Several nondestructive assay techniques, e.g., calorimetry and spontaneous fission assay detectors, require a knowledge of plutonium and americium isotopic ratios to convert their raw data to total grams of plutonium. This paper describes a nondestructive technique for calculating plutonium-238, plutonium-240, plutonium-241 and americium-241 relative to plutonium-239 from measured peak areas in the high resolution gamma-ray spectra of solid plutonium samples. Gamma-ray attenuation effects have been minimized by selecting sets of neighboring peaks in the spectrum whose components are due to the different isotopes. Since the detector efficiencies are approximately the same for adjacent peaks, the accuracy of the isotopic ratios are dependent on the half-lives, branching intensities and measured peak areas. The data presented describes the results obtained by analyzing gamma-ray spectra in the energy region from 120 to 709 keV. The majority of the data analyzed was obtained from plutonium material containing 6% plutonium-240. Sample weights varied from 0.25 g to approximately 1.2 kg. The methods have also been applied to plutonium samples containing up to 23% plutonium-240 with weights of 0.25 to 200 g. Results obtained by gamma-ray spectroscopy are compared to chemical analyses of aliquots taken from the bulk samples.

Nondestructive assay (NDA) is important in safeguarding plutonium since it provides the means to measure all of the feed, product and scrap material generated in the fuel fabrication process. Calorimetry and spontaneous fission detection are two important NDA techniques which can be used to account for plutonium. In order to convert the raw data from these measurements to grams of plutonium, the relative isotopic composition of the samples must be known. This isotopic ratio data can be provided by traditional chemical methods, gamma-ray analysis of solutions or gamma-ray analysis of the bulk sample itself.

Gamma-ray analysis of the bulk sample is attractive because it allows a totally nondestructive measurement. We have taken data principally on three size containers: 1) gallon cans containing 20 to 400 g of plutonium; 2) quart cans containing between 10 and 2500 g of plutonium; and 3) analytical vials containing 1 to 10 g of plutonium. The sample categories include plutonium oxide, plutonium metal, scrap, incinerator ash, ash heels, fluorides, crucibles, slag, scarfings, green cake and mixed plutonium-uranium oxides.

The desired accuracy for all the isotopic ratios is less than 3%. With these uncertainties and a calorimetric uncertainty of 0.25%, the total uncertainty in the plutonium analysis is less than 1%.

*Mound Laboratory is operated by Monsanto Research Corporation for the U. S. Energy Research and Development Administration under contract No. E-33-1-GEN-53.

The basis for the nondestructive measurement of plutonium isotopic ratios using gamma-ray spectroscopy involves the analysis of a spectral group whose members belong to different isotopes. Ratios of the areas of neighboring gamma-ray peaks are related to the isotopic abundance ratios by the expression

$$\frac{I_i}{I_j} = \frac{N_i \lambda_i B_i \varepsilon_i S_i}{N_j \lambda_j B_j \varepsilon_j S_j}$$

where I, N, λ, B, ε, and S are the measured peak area, the number of nuclei, the nuclear decay constant, the absolute branching intensity, the detector efficiency and the self-absorption for the gamma rays with energies E_i and E_j from isotopes i and j, respectively.

The ratios of the relative efficiencies, $(\varepsilon_i S_i)/(\varepsilon_j S_j)$, is assumed to be 1 for gamma rays whose energies differ by less than 10 keV. For the bulk samples, the further assumption is made that the plutonium isotopic composition is constant throughout the volume of the sample.

A high resolution Ge(Li) detector is used to acquire the gamma-ray spectra. The sample is rotated while acquiring data to reduce the effects of inhomogeneities due to the distribution of plutonium in the sample. A four-inch thick lead collimator with a 0.25" wide vertical slit is used to reduce the response to material located near the edge of the container. This allows the detector essentially to view the sample only along a diameter and improves the signal-to-background ratio by absorbing photons which are Compton scattered in the sample. A 0.030" (0.8 mm) cadmium absorber is used to further reduce the effects of low energy gamma rays. The data for all the isotopic ratios is measured with one detector.

Figure 1 shows the spectrum of a plutonium sample containing 94% plutonium-239 and 6% plutonium-240. The prominent gamma rays from each isotope of interest are indicated.

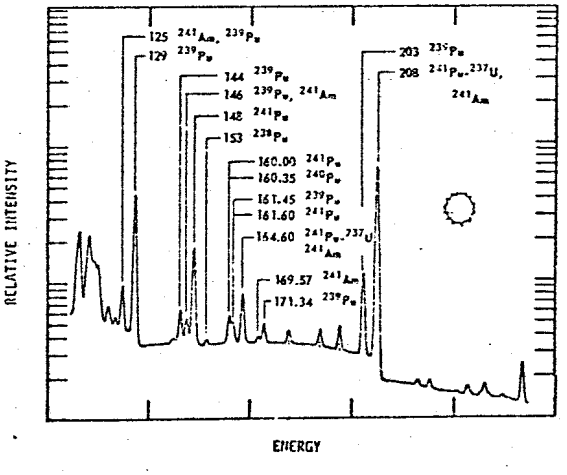

Fig. 1A - Gamma-ray Spectrum of Plutonium Sample Containing 94% Pu-239 Showing Energy Region from 120 to 220 keV.

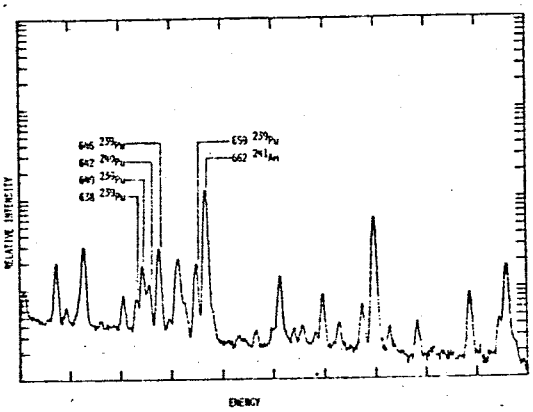

Fig 1B - Gamma-ray Spectrum of Plutonium Sample Containing 94% Pu-239 Showing 600 keV Energy Region.

The gamma rays used for each isotopic ratio are shown in Table 1. Also listed in the table are the conversion factors calculated using the branching intensities and half-life values of Gunnink and Morrow[1]. Peak areas are extracted using the programs GAUSS V[2] or GRPANL[3]. Both are nonlinear least squares fitting routines. The former runs on an IBM-360 computer; the latter on PDP-8 computers. The program used is dependent upon the spectroscopy system employed in the acquisition of the spectrum. Both programs give equally valid results when applied to the same spectrum.

TABLE 1

GAMMA-RAYS AND CONVERSION FACTORS USED FOR ISOTOPIC DETERMINATIONS

Isotopic Ratio	Gamma-rays	Conversion Factors to Express Ratio in ppm
$\frac{^{238}Pu}{^{239}Pu}$	$\frac{153\ keV}{144\ keV}$	$1010\ \frac{I_{153}}{I_{144}}$
$\frac{^{238}Pu}{^{241}Pu}$	$\frac{153\ keV}{148\ keV}$	$1.185\ \frac{I_{153}}{I_{148}}$
$\frac{^{240}Pu}{^{239}Pu}$	$\frac{642\ keV}{640\ keV}$	$148300\ \frac{I_{642}}{I_{640}}$
$\frac{^{240}Pu}{^{241}Pu}$	$\frac{160.35\ keV}{164.6\ keV}$	$50.7\ \frac{I_{160}}{I_{165}}$
$\frac{^{241}Pu}{^{239}Pu}$	$\frac{208\ keV}{203\ keV}$	$623.7\ \frac{I_{208}}{I_{203}}$
$\frac{^{241}Pu}{^{239}Pu}$	$\frac{148\ keV}{144\ keV}$	$852.7\ \frac{I_{148}}{I_{144}}$
$\frac{^{241}Am}{^{239}Pu}$	$\frac{662\ keV}{659\ keV}$	$487.5\ \frac{I_{662}}{I_{659}}$

The accuracy of nondestructive techniques can be established by comparing the results on aliquots with their subsequent chemical analysis.

Over the past three years, we have compared the nondestructive (NDA) and chemical analysis (DA) results for forty-five 9 g plutonium metal samples (93% plutonium-239). The average percent differences $\left(\frac{NDA-DA}{DA}\right)$ are: plutonium-238 (1.0 ± 11.7)%; plutonium-240 (-5.2 + 6.4)%; plutonium-241 (4.2 ± 5.7)% and americium-241 (-2.9 ± 4.9)%.

The majority of the bulk samples investigated have been 93% plutonium-239. They are sent to Mound Laboratory for calorimetric assay as a part of a plutonium verification program[4]. Analytical aliquots are chosen from selected samples prior to shipment to Mound Laboratory. A combination of chemical and nondestructive measurements of the isotopic ratio is used with the calorimetry data to give an uncertainty in the plutonium content of less than 1%. Because of the overall time commitments of the program, samples can be counted for only four hours. In the most recent sample exchange, 20 aliquots and 79 samples were processed. The percent differences for the aliquots $\left(\frac{NDA-DA}{DA}\right)$ are: plutonium-238 (-1 ± 7)%; plutonium-240 (-7 + 11)%; plutonium-241 (-2 + 3)% and americium-241 (1 ± 7)%. When the isotopic ratios of the aliquots (A) are compared to the cans (C) from which they were drawn, the average percent differences $\frac{(C-A)}{A}$ are: plutonium-238 (1 ± 14)%; plutonium-240 (1 + 18)%; plutonium-241 (1±2)%; americium-241 (1± 7)%. If we use uncertainties of 15% for $^{238}Pu/^{239}Pu$, 15% for $^{240}Pu/^{239}Pu$, 3% $^{241}Pu/^{239}Pu$ and 7% $^{241}Am/^{239}Pu$, the resulting uncertainty in the calorimetric assay would be 3%.

We are presently extending these techniques to lower fissile material. As a final example of our results, we analyzed five reactor fuel pins containing 86% plutonium-239. Comparison of the gamma-ray analysis of the pins with chemical analysis of the fuel at the time of fabrication yields the following differences: plutonium-238 (5.2 ± 6.5)%; plutonium-240 (-0.8 ± 5.8)%; plutonium-241 (0.2 ± 1.4)%; americium-241 (-5.1 ± 2.4)%. The use of gamma-ray isotopics in a calorimetric assay would yield total plutonium values which are 0.43% higher than corresponding values using chemical isotopics.

The technique we have developed in general meets the goals we have established for the program[5]. We have applied it to small samples with promising results as indicated by the results of the plutonium metal and fuel pin data. We feel it also shows promise for application to bulk samples as indicated by the verification program results. We use it routinely for plutonium-241 and americium-241 isotopic data and to verify that the aliquot is representative of the bulk sample. The results do not meet the 3% accuracy we set for the plutonium-238 and plutonium-240 isotopics. Plutonium-238 fails as a result of the weak intensity of the 153 keV peak at the 100 ppm level of plutonium-238. The plutonium-240 fails because of the low intensity of the 640 keV peaks and the requirement that all isotopic data be obtained in four-hour data acquisition time. We are guardedly optimistic that using a two detector system, one to examine the region up to 210 keV and the second the region at 600 keV, this goal can be achieved. These efforts are presently underway.

REFERENCES

1. R. Gunnink and R. J. Morrow, "Gamma-ray Energies and Absolute Branching Intensities for $^{238,239,240,241}Pu$ and ^{241}Am", UCRL-51087, University of California Radiation Laboratory, Livermore, California, July 22, 1971.

2. R. G. Helmer and M. H. Putnam, "GAUSS V: A Computer Program for the Analysis of Gamma-ray Spectra from Ge(Li) Spectrometers", ANCR-1043, Aerojet Nuclear Corporation, Idaho Falls, Idaho (January, 1972).

3. R. Gunnink, "A System for Plutonium Analysis by Gamma-ray Spectrometry - Part 2: Computer Programs for Data Reduction and Interpretation", UCRL-51577 Part 2 University of California Radiation Laboratory, Livermore, California (April 18, 1974).

4. Raymond S. George and Roy B. Crouch, "Inspector Measurement Verification Activities", Proceedings of the Sixteenth Annual Meeting of the INMM, June, 1975, p. 327-336.

5. Nuclear Safeguards Progress Reports, Mound Laboratory, MLM-2302 (April, 1976), MLM-2286 (December, 1975), MLM-2185 (December, 1974).