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FUEL ROD ENRICHMENT DETERMINATION BY GAMMA-RAY SPECTROMETRY^(a)

MASTER

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ABSTRACT

The plutonium concentrations in mixed oxide fuel rods containing distinct sizes of particulate plutonium-oxide have been determined using high resolution gamma-ray spectrometry. The fuel rod enrichments were in the range 2 to 2.5 wt.% PuO_2 in natural UO_2 . Individual rod enrichments were determined to within $\pm 5\%$ by comparison with a known standard. Differences in plutonium isotopic composition were discernable also, in addition the differences in plutonium enrichment. The technique provides an inexpensive, non-destructive method of determining in-situ plutonium composition and content of assembled fuel rods, for purposes of quality assurance and accountability control.

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INTRODUCTION AND SUMMARY

One phase of the Plutonium Utilization Program, conducted for the USAEC by Battelle-Northwest, has been an investigation of the variation of the neutronic parameters ⁽¹⁾ of test lattices with respect to different sizes of PuO_2 particles in the UO_2 - PuO_2 fuel mixture. Ten sets of four rods each were fabricated to a specified enrichment of 2 wt.% PuO_2 in natural UO_2 , with plutonium particle sizes ranging from zero microns to approximately 350 microns in diameter. The rod sets were used in experiments performed in the Physical Constants Testing Reactor (PCTR) to determine k_∞ of the test lattice as a function of the diameter of the PuO_2 particles. The reactivity worths of the different rod sets measured in the PCTR, necessary to the determination of k_∞ , were not consistent with the predicted reactivity variations due to the differences in particle size and it was suspected that the enrichments of the rods were not truly 2 wt.% PuO_2 , as specified.

The results from a destructive analysis of four pellets, two taken from a solid solution rod (zero micron particles), and two from a rod containing 350 micron particles, indicated that the pellets containing the particles could be over-enriched as much as 20 percent. Therefore, one rod from each set was gamma scanned using a high resolution lithium-drifted germanium detector by which the intensities of the photo-peaks for ^{239}Pu , ^{241}Am , ^{241}Pu and ^{237}U were determined. A comparison of specific photo-peak areas from each particle rod with similar photo-peak areas from

the solid solution rods was used to determine the enrichment of the particle rods.

The photo peak area ratios were not directly related to the enrichment ratios until the data had been corrected for the variations in the gamma-ray attenuation factors with respect to the diameter of the PuO_2 particle. The corrected gamma-ray measurements were in agreement with the destructive measurements made on the two pellets containing the 350 micron particles. On the basis of these normalization points it was possible to determine the enrichment of all ten rod sets from the gamma-ray photo peak area ratios.

Description of the Mixed Oxide Fuel Rods

A complete list of the various fuel rod sets which were manufactured for the PCTR experiment is presented in Table I. Originally, five rod sets were prepared with nominal particle diameters of zero microns (called solid solution rods), 50 microns, 100 microns, 200 microns, and 350 microns. Additional sets of rods with similar particle diameters were made as different particle manufacturing techniques were developed. All the rods were 20 inches in length, with the fuel contained in zirconium tubing, 0.565 inches O.D. and 0.030 inch wall thickness.

Discovery of the Problem

The particle rods were initially suspected to be of different enrichments on the basis of reactivity measurements made with the rods in the Physical Constants Testing Reactor. The results of these measurements are presented in Table I. The fuel rods which contain the 350 micron particles were almost as reactive as the solid solution rods. Foil measurements⁽²⁾

TABLE I

REACTIVITY WORTH OF A TEST ROD IN THE
CENTER OF THE PCTR TEST CAVITY

<u>Rod Identification</u>	<u># Rods In Set</u>	<u>Rod Type</u>	<u>(grams/cc) UO₂-PuO₂ Density</u>	<u>(¢) Reactivity Worth</u>
8SC	4	Solid Solution Ball Milled (Manufactured from same material as 8RA350)	8.704	37.8 ± 0.3
8BMB	4	Solid Solution Ball Milled (Manufactured from same material as 8B rods)	8.571	38.2 ± 0.3
8BME	1	Solid Solution Ball Milled (Last rod Manufactured)	8.474	38.1 ± 0.3
8P50	4	50 Micron Induction Plasma	8.601	39.1 ± 0.3
8B100	4	100 Micron Jet Milled	8.739	39.3 ± 0.3
8P100	4	100 Micron Induction Plasma	8.638	39.0 ± 0.3
8B200	4	200 Micron Jet Milled	8.718	38.3 ± 0.3
8P200	4	200 Micron Induction Plasma	8.657	38.3 ± 0.3
8RA350	2	350 Micron Jet Milled-Vibration Tested for Roundness	8.798	37.4 ± 0.3
8B350	4	350 Micron Jet Milled	8.691	37.8 ± 0.3

of the self shielding effects in the PuO_2 particles indicated the 350 micron rod sets should have been approximately 6% less reactive than the solid solution rods. The foil measurements and the reactivity measurements were repeated and the same results obtained, confirming the suspicion that the rod enrichments were not as specified. Thereupon, one of the solid solution rods and one of the 350 micron particle fuel rods (both manufactured at approximately the same time) were opened and two fuel pellets were removed from each rod. The four pellets were dissolved separately and analyzed for their plutonium content.

The results of the chemistry analysis ⁽³⁾ are listed in Table II. It is apparent from the chemistry results that there could be as much as twenty percent difference between rods.

Development of the Analysis Method

Upon receiving confirmation of the suspected enrichment variations in the fuel rods, a non-destructive analysis was devised to determine the enrichment variations in all the rods using gamma-ray measurement. The intensity, I_{ij} , of a gamma-ray associated with the i^{th} isotope of plutonium in the j^{th} fuel rod can be expressed by:

$$I_{ij} = N_{ij} \lambda_i \gamma_i \epsilon_i = \frac{N_0}{A_i} (\text{wt}\%)_j \rho_j (\text{IF})_{ij} \lambda_i \gamma_i \epsilon_i \quad (1)$$

where λ , γ , and ϵ are the decay constant, gamma-rays per disintegration and detector efficiency respectively. N_0 is Avogadro's number, A_i is the atomic weight, $(\text{wt}\%)_j$ is the percent by weight of plutonium in the $\text{UO}_2 - \text{PuO}_2$ mixture, ρ_j is the density of the mixture, and $(\text{IF})_{ij}$ is the isotopic fraction $\text{Pu}^i / \text{Pu}^{\text{TOTAL}}$ in the j^{th} rod.

TABLE II

RESULTS FROM ANALYSIS OF UO_2 - PuO_2 FUEL ROD SPECIMENS

Rod Type Pellet #	8BMB Solid Solution		8B350 350 Micron Particles	
	#1	#2	#1*	#2
Pu/U (wt. ratio)	0.0202 ±.0001	0.0202 ±.0001	0.0229 ±.00015	0.0249 ±.00015
wt.% Pu (Pu + U)	0.0198 ±.0001	0.0198 ±.0001	0.0224 ±.00015	0.0243 ±.00015

Average Pu Isotopic Composition (All samples agreed within accuracy of measurement)

Pu-238 wt.% (Pu)	0.0340	±0.0035
Pu-239 wt.% (Pu)	90.8524	±0.0200
Pu-240 wt.% (Pu)	7.9847	±0.0200
Pu-241 wt.% (Pu)	1.0687	±0.0050
Pu-242 wt.% (Pu)	0.0602	±0.0018

U Isotopic Composition

U-234 wt.% (U)	0.0054	±0.0001
U-235 wt.% (U)	0.7138	±0.0002
U-238 wt.% (U)	99.2808	±0.0002

Date of Analysis: March 1970

*dissolution difficulties, result questionable

For the low energy gamma rays being studied here, the fuel rod appears as if it were an infinite slab, with infinite thickness, and a uniformly distributed source. The photo-peak intensity at the surface of the rod can then be expressed by: (4)

$$I_{ij} = \frac{N_{ij} \lambda_{ij} \gamma_i \epsilon_i}{2\mu_j \rho_j} = \frac{N_o (\text{wt}\%)_j (IF)_{ij} \lambda_{ij} \gamma_i \epsilon_i}{A_j 2\mu_j} \quad (2)$$

where μ_j is the mass attenuation coefficient of the $\text{UO}_2 - \text{PuO}_2$ mixture. By constructing ratios of intensities of the i^{th} photo-peak from the unknown j^{th} rod and from the known standard rods, the percent by weight of plutonium in the j^{th} rod is obtained.

$$(\text{wt}\%)_j = \frac{I_{ij}}{I_{is}} (\text{wt}\%)_s \frac{(IF)_{is}}{(IF)_{ij}} \quad (3)$$

This expression is valid for $\text{UO}_2 - \text{PuO}_2$ mixtures in which the plutonium is very finely divided (essentially zero microns particle diameter) and is distributed homogeneously throughout the UO_2 diluent.

In the case of the finite-sized plutonium particles, a correction is needed to account for the increased absorption of the gamma-rays in the PuO_2 microspheres as compared with an equivalent microsphere of the $\text{UO}_2 - \text{PuO}_2$ mixture. The ratio of the photo-peak intensity at the surface of a $\text{UO}_2 - \text{PuO}_2$ microsphere to the photo-peak intensity at the surface of the PuO_2 microsphere is given by: (5)

$$C_i(R) = \frac{\frac{S_v}{2\mu_2 \rho} \left\{ 1 - \frac{1}{2\mu_2 \rho_2 R} + \frac{e^{-2\mu_2 \rho_2 R}}{2\mu_2 \rho_2 R} \right\}}{\frac{S_v}{2\mu_1 \rho} \left\{ 1 - \frac{1}{2\mu_1 \rho_1 R} + \frac{e^{-2\mu_1 \rho_1 R}}{2\mu_1 \rho_1 R} \right\}} \quad (4)$$

where the subscripts 1 and 2 refer to PuO_2 at a density of 11.47 g/cm^3 and $\text{UO}_2 - \text{PuO}_2$ at a density of 8.6 g/cm^3 respectively. R is the radius of the microsphere. The volume sources, S_v , are assumed to be equal and cancel in the ratio. The values of $C_i(R)$ are illustrated in Figure 1, for the range of microsphere radii and photo-peak energies of interest in these measurements.

Upon applying the above correction to the measured photo-peak intensities, equation (3) becomes:

$$(\text{wt}\%)_j = \frac{I_{ij} C_i(R) (\text{wt}\%)_s (\text{IF})_{is}}{I_{is} (\text{IF})_{ij}} \left(\frac{\mu_{ij}}{\mu_{is}} \right) \quad (5)$$

Each photo-peak from the j^{th} rod provides an estimate of the unknown weight percent, $(\text{wt}\%)_j$. Therefore, the best estimate is given by:

$$(\text{wt}\%)_j = \frac{(\text{wt}\%)_s}{N} \sum_{i=1}^N \frac{I_{ij} C_i(R)}{I_{is}} \frac{(\text{IF})_{is}}{(\text{IF})_{ij}} \left(\frac{\mu_{ij}}{\mu_{is}} \right) \quad (6)$$

In some instances the actual isotopic fractions may not be well known, especially in the case of ^{241}Pu . Unless the date of the isotopic determination is known, the ^{241}Pu content will not be known, even though the ^{239}Pu and ^{240}Pu contents will not have changed significantly. Using the ^{241}Pu isotopic fraction measured at an earlier (and unknown) time will result in a biased estimate of $(\text{wt}\%)_j$. An estimate of the $^{239}\text{Pu}/^{241}\text{Pu}$ isotopic ratio present in the fuel at the time of the gamma-ray measurements can be obtained by constructing ratios of photo-peak intensities from the i^{th} isotope (^{239}Pu) to the k^{th} isotope (^{241}Pu), using equation (5). The isotopic fraction of the k^{th} isotope is then given by:

FIGURE 1. CORRECTION FACTOR FOR SELF-ABSORPTION OF GAMMA-RAYS IN PuO_2 MICROSPHERES AS A FUNCTION OF MICROSPHERE DIAMETER, WITH GAMMA-RAY ENERGY AS A PARAMETER

$$(IF)_{kj} = (IF)_{ij} \frac{(IF)_{ks}}{(IF)_{is}} \cdot \frac{I_{kj} C_k(R)}{I_{ij} C_i(R)} \cdot \frac{I_{is}}{I_{ks}} \left(\frac{\mu_{kj} \mu_{is}}{\mu_{ks} \mu_{ij}} \right) \quad (7)$$

Equations (6) and (7) have been used, together with the isotopic data in Table II and the photo-peak intensity data in Table III to determine the actual percent by weight of plutonium present in the different sets of rods.

Experimental Procedure

The scanning system used in the measurement program is illustrated in Figure 2. A high resolution lithium-drifted germanium (Ge-Li) planar diode (1 cm x 3 cm x 3 cm) viewed the fuel rod being scanned through an aperture 0.25 inches x 0.625 inches. Each rod was rotated about its longitudinal axis at 1 RPM during scanning to average any localized enrichment abnormalities around the circumference of the rod.

The data were collected using a multi-channel pulse height analyzer, with an energy width of 0.49 keV per channel. Only photo-peaks having energies less than 250 keV were examined. The gamma-ray energy spectrum obtained from Rod 8BMB, a solid solution rod, is shown in Figure 3, with the photo-peaks used in the enrichment analysis identified. The first and last channels included in the summation of counts used to determine the photo-peak areas were:

$^{241}\text{Am} + ^{237}\text{U}$	(59.5 keV)*	Channels 110 to 133
^{239}Pu	(129.3 keV)*	Channels 256 to 267
^{241}Pu	(148.6 keV)*	Channels 296 to 305
^{237}U	(208.0 keV)*	Channels 414 to 424

*Reference 7

TABLE III

RESULTS OF ANALYSIS OF GAMMA SCAN DATA ON PARTICLE FUEL RODS

<u>Rod Number</u>	<u>Number of Scan Points</u>	<u>Average Number of Counts</u>			
		<u>59.3 keV Peak</u>	<u>129.3 keV Peak</u>	<u>148.6 keV Peak</u>	<u>208.0 keV Peak</u>
8BMB*	10	747780	8845	6491	33418
8SC	10	706267	9203	4269	22373
8BME	3	872343	9410	8400	43405
8P50	10	814458	10055	7425	38646
8P100	3	826287	10529	8060	41106
8B100	10	883852	10788	7888	40390
8P200	3	768842	10094	7689	40795
8B200	10	761653	9925	7459	39595
8B350	10	730002	9869	7443	40234
8RA350	10	701047	10218	4685	25719

*Solid solution rod used as standard

FIGURE 2. Ge-Li Detector System to Determine Enrichment in Mixed Oxide Fuel Rods

FIGURE 3. Measured Gamma Ray Photopeaks from Mixed Oxide Fuel Rod 8BMB

The average number of background counts per channel was derived by averaging the counts in the four channels adjacent to the first and last channels included in the peak.

One sample rod was chosen from each set of rods listed in Table I. It was assumed that the fuel enrichment in any single rod of a four rod set was the same as that in the other rods of the set. This assumption was partially tested during the PCTR reactivity measurements in which single rods of a particular particle size and manufacturing technique were calibrated against the other three rods in the same set and no significant reactivity differences were measured ($\leq 0.1\%$ variation).

Initially, all rods were scanned at a minimum of three axial locations, 4.425, 9.425, and 14.425 inches from the end of rod, which placed the aperture at the centers of three separate pellets in the rod. After three measurements were taken on each rod, additional measurements were made as time permitted by starting at 2.425 inches and moving the scanning point at two inch intervals until 10 separate measurements had been completed.

A summary of the photo-peak intensity data is presented in Table III. The tabulated counts for each peak do not include the background and have been normalized to an equivalent live-time counting interval.

A review of these data and of the literature for gamma ray emission leads to several conclusions:

- The 59.5 keV photo-peak is contaminated with other low energy gamma-rays and is not a good choice as an enrichment indicator.
- Rods 8SC and 8RA350 have similar isotopic ratios but have lower $^{241}\text{Pu}/^{239}\text{Pu}$ ratios than the other rod sets.

- Rod 8BME appears to have a higher 241/239 ratio than the other rods. However, only three scans were made on this rod and the result is not statistically conclusive.
- All other rods have the same 241/239 ratios within the measurement uncertainty ($\pm 3\%$) and therefore can be assumed to have the same isotopic fractions.
- The ratios of peak areas between rods thought to contain the same plutonium isotopic fractions indicate that the attenuation coefficient for each gamma-ray from plutonium varies as a function of the diameter of the plutonium particle.

Experimental Results

The data presented in Table III, after correction for particle attenuation effects, were used in the relationships derived earlier to determine the plutonium concentrations in the various rods. Rod 8BMB was selected as the standard because (1) being a solid solution rod, no particle attenuation effects were present, and (2) an accurate chemical and isotopic analysis of pellets from the 8BMB rod set was available.

The results of this analysis are presented in Table IV. It is worthwhile to note that while Rod 8BMB was used as the standard rod in the analysis, the enrichment derived for Rod 8B350 agrees with the enrichment determined by destructive analysis of pellets from the 8B350 rod set to well within the measurement uncertainty, thus increasing confidence in the derived concentrations where no cross checks were available.

TABLE IV

ENRICHMENT OF $\text{UO}_2\text{-PuO}_2$ PARTICLE FUEL DETERMINED BY
GAMMA SCANNING, CHEMISTRY, AND REACTIVITY MEASUREMENTS

<u>Rod Designator</u>	<u>Rod Type</u>	<u>wt.% PuO_2 (by gamma scanning)</u>	<u>*Relative Reactivity Worth in PCTR</u>	<u>Chemical Analysis of Two Pellets (wt.% PuO_2)</u>	<u>Fuel Mixture Density (g/cc)</u>
8BMB	Solid Solution	Used as Base (1.98)	1.0	1.98 ± 0.01	8.571
8SC	Solid Solution	2.08 ± 0.08	1.043 ± 0.007	None	8.704
8BME	Solid Solution	2.10 ± 0.09	1.018 ± 0.007	None	8.474
8P50	50 Micron	2.29 ± 0.09	1.088 ± 0.007	None	8.601
8P100	100 Micron	2.47 ± 0.10	1.074 ± 0.007	None	8.638
8B100	100 Micron	2.48 ± 0.10	1.102 ± 0.007	None	8.739
8P200	200 Micron	2.41 ± 0.08	1.030 ± 0.007	None	8.657
8B200	200 Micron	2.35 ± 0.09	1.036 ± 0.007	None	8.718
8RA350	350 Micron	2.47 ± 0.09	0.997 ± 0.007	None	8.798
8B350	350 Micron	2.42 ± 0.09	0.981 ± 0.007	2.43 ± 0.015	8.691

*Note: The reactivity measurements are a function of the self shielding effects of the particles and the fuel mix densities as well as the amount of enrichment.

Conclusions

Several interesting conclusions can be drawn from the results of this study.

- It appears that the rod sets were manufactured from three different batches of plutonium. The differences in isotopic ratios coincide with the different dates and methods of production. Rod sets 8SC and 8RA350 were made with one batch of plutonium, rod sets 8BMB, 8P50, 8B100, 8P200, 8B200 and 8B350 were made from a second batch of plutonium, and rod sets 8BME may have been made from a third batch of plutonium, each batch having a different 239/241 ratio.
- The enrichments of the various particle size rod sets differed significantly from the design value of 2.0 wt% PuO_2 in UO_2 , with values ranging from ~ 2 wt% to as much as 2.50 wt%.
- The enrichments of the rod sets could be determined with a statistical uncertainty of less than 5% (one standard deviation) of the measured value.

The technique described here has been shown to be a valuable and useful method of assaying the plutonium content of unirradiated mixed oxide fuel rods. The method permits comparison of enrichments, either microscopic (pellet by pellet) or macroscopic (rod by rod), of manufactured rods without requiring destructive analysis of the rods, provided that a suitably characterized standard rod is available, thus providing an inexpensive and relatively rapid means of quality assurance and accountability control.

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