Intrinsic Densitometry—In-Plant Evaluation

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INTRINSIC DENSITOMETRY—IN-PLANT EVALUATION

by

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ABSTRACT

This report describes the testing of the intrinsic densitometry (ID) technique for in-plant applications. We found that the ID method can determine the plutonium concentrations to between 2 and 3% at concentrations of 100 g/l to 200 g/l with quartz cells and a measurement time of 3600 s. The precision can be improved to 1 to 2% with a higher counting rate. We also found that nitric acid concentration and the impurity level of uranium in the product plutonium solution do not affect the concentration measurement. When this technique is applied to plutonium solutions in stainless steel pipes, we found that similar precision in plutonium concentration can be achieved using a high-count-rate detector. The precision, however, is reduced with aged plutonium solutions.

I. INTRODUCTION

A measurement of the plutonium concentration in a sample is always necessary for nuclear material control and accounting. This report describes in-plant experiments with the intrinsic densitometry (ID) method of determining plutonium concentration (and isotopic distribution) that requires *no* external radioactive sources or x-ray generators but relies only on natural radiation. The method is ideally suited to assay reasonably pure plutonium solutions, such as the eluate solutions from plutonium recovery and the product solutions of reprocessing plants. The method can be applied to aged or freshly separated plutonium and can be used to measure plutonium concentrations in pipes or tanks.

This paper describes in-plant experiments for determining the plutonium concentration from the gamma rays and x-rays emitted by plutonium without relying on external sources. We will discuss the experiments with this method with high-burnup samples and also measurements with different sample thicknesses and impurities. We will also discuss experiments with plutonium solution in pipes. The goal of these experiments is to explore the application of this technique to monitoring concentrations in pipes and tanks in plutonium processing plants.

This project was carried out under the cooperative agreement between the Power Reactor & Nuclear Fuel Development Corporation (PNC) and the DOE in research and development concerning materials control and accounting for safeguards. The measurements were carried out at the Tokai Reprocessing Plant at Tokai, Japan.

II. PRINCIPLE

The principle of ID is based on the fact that the K-absorption edge for plutonium is at 121.8 keV. If we select one peak from the plutonium spectrum on both sides of the edge, the two peaks will undergo different amounts of attenuation through the solution. The dependence can be described in functional form if the far-field approximation can be applied. For a narrow concentration range, the plutonium concentration is a linear function of the ratio of peak areas. The principle of the technique is discussed in Refs. 1 and 2 for low-burnup and high-burnup plutonium respectively.

For high-burnup solutions, the two peaks selected are the 111-keV x-ray from plutonium and the 148-keV peak from ^{241}Pu decay. The μ values for plutonium at these two energies differ by 1.29 cm^2/g ; because of the difference in the μ values, the ratio of the 111/148 peak intensities is a function of the plutonium concentration. The peak areas of the 111-keV and 148-keV photo peaks were determined by the MGA2 isotopic program,³ which also determines the isotopic distribution. The ratio of the 111-keV and 148-keV peak areas is expressed as

$$\frac{I(111)}{I(148)} = kCF(\text{ISO}) * \frac{CF(\text{AT})_{148}}{CF(\text{AT})_{111}}$$

where the $CF(\text{AT})$ is the correction factor for attenuation and $CF(\text{ISO})$ is the isotopic correction factor. The isotopic factor is discussed in Ref. 2. For high-burnup plutonium, the isotopic factor² is as follows:

$$\text{Isotopic factor} = 1 + \frac{f_8}{f_1} * \frac{x_8}{x_1} + \frac{f_9}{f_1} * \frac{x_9}{x_1} + \frac{f_0}{f_1} * \frac{x_0}{x_1}$$

where f_i = weight fractions of various plutonium isotopes
and x_i = x-ray/(s • g) (at 111 keV) of i plutonium isotopes.

III. EXPERIMENTAL SETUP—PART 1

The experiment is separated into two parts. In the first part of the experiment, the plutonium solutions are contained in quartz cells; in the second part of the experiment, the plutonium solution is contained in stainless steel pipes.

For the first part of the experiment, the plutonium solutions were in quartz cells inside a glove box; the cell was placed in a lead shield to reduce the background radiation. The gamma rays from the solution were collimated by a lead collimator 3 cm long with a hole 1 cm in diameter. The detector had a 2-cm³-active-volume, side-looking Low Energy Photon Spectrometer (LEPS). The signal from the detector is processed by a linear amplifier and stored by a Canberra Industries S100 multichannel analyzer. Each sample was assayed for 3600 s of live time. The experimental setup is shown in Fig. 1 below.

IV. EXPERIMENTAL RESULTS

A. Plutonium Concentration and Peak Ratio

Three different concentrations of high-burnup plutonium solutions were prepared with concentrations ranging from 100 to 208 g/l. The concentrations were determined by titration, which in general has a precision of 0.1%. Two batches of plutonium with different isotopic mixes were prepared. The isotopic distributions are shown in Table I.

Fig. 1. Experimental setup for the ID measurement. The quartz cells are placed in lead shields inside a glove box.

Table I. Isotopic distribution of plutonium samples				
	Sample A (wt%)		Sample B (wt%)	
	MGA2	Mass spec.	MGA2	Mass spec.
²³⁸ Pu	1.2	1.1	1.3	1.3
²³⁹ Pu	62.2	62.7	58.0	57.0
²⁴⁰ Pu	24.3	23.8	26.4	27.8
²⁴¹ Pu	9.2	9.0	10.1	10.5
²⁴² Pu	3.1	3.7	4.2	3.4

Figure 2 shows the plutonium concentration plotted versus the 111/148-keV peak ratios, corrected by the isotopic factor. The data from sample B are also shown in Fig. 2. The concentrations plotted versus the corrected ratios can be fitted with a straight line; the correlation factor was 0.99. The data from sample B agreed with the results from sample A to within 1%. This indicates that the ID method can be used to determine plutonium concentration and this method can be applied to solutions having different isotopic distributions.

B. Repeatability

The precision of the peak ratio method (111/148 keV) was measured on repeated runs. The sample used for this measurement had a concentration of 208 g/l. The measurement was repeated ten times for the solution. The data for repeatability are shown in Table II below.

Fig. 2. Plutonium concentration as a function of the measured 111-keV/148-keV ratios after the isotopic correction. Plutonium solution in quartz cells. Sample A and sample B have different isotopic distributions.

Table II. Precision of repeated runs		
111-keV peak area	148-keV peak area	111/148 ratio
72290	71301	1.013
72045	70741	1.018
71177	71143	1.000
70540	70520	1.000
70496	71152	0.991
71610	71226	1.005
71528	71107	1.005
71031	71084	0.999
71836	71523	1.004
72290	71301	1.014
	Average	1.005
	SD	0.8%

The repeatability or precision of the ratio is 0.8% for the present experiment. The precision of the plutonium concentration determination is approximately 4%, according to the sensitivity factor in Ref. 2 for a 2-cm cell. It should be pointed out that the precision can be improved by using higher counting rates and also longer counting times; the present counting rate of ~1000 cps can be easily increased by a factor of 10.

C. Effects of Impurities

Because 111 keV and 148 keV are sufficiently apart in energy, the effects of impurities on the ID method may not be negligible. The effects of nitric acid and uranium, which are often contained as an impurity in the plutonium product solution, were studied.

In this experiment, we kept the plutonium concentration constant at 166 g/l. The data are summarized in Table III. The nitric acid molarity variation from 1.5 M to 3.2 M to 5 M had no measurable effect on the determination of the plutonium concentration. In general, a low-Z matrix has a negligible effect on gamma-ray attenuation.

In the next experiment, we used the same plutonium concentration (166 g/l) but varied the uranium concentration from 0.3 g/l to 35 g/l. The first concentration represents the upper limit of the uranium impurity level in plutonium; the second represents the possible uranium level in mixed solution. The data are summarized in Table IV. The results suggest that the concentration of uranium at the impurity level (<0.3 g/l) interferes little with the plutonium concentration determination. The presence of a large amount of uranium, however, has a measurable effect on the plutonium concentration. In fact, the total concentration determined is close to the sum of the uranium and plutonium concentrations.

D. Sample Thickness Effect

We studied the effect of sample thickness on the concentration and isotopic determination. Four types of cells with thicknesses of 1, 2, 3, and 5 cm were measured with an experimental setup similar to the one shown in Fig. 1. The concentration of the sample that we used is

Table III. Effect of nitric acid on plutonium concentration	
Nitric Acid	Measured Value (g/l)
1.5 M	166.9 ± 2.1
3.2 M	165.2 ± 1.9
5.0 M	166.2 ± 2.3

Table IV. Effect of presence of uranium on the plutonium concentration determination	
U Conc. (g/l)	Pu Conc. (g/l) Measured
35	204.8 ± 1.5
0.3	165.4 ± 1.6

~210 g/l. A plot of the peak ratio of the 111-keV and 148-keV peak areas versus the sample thickness is shown in Fig. 3. The optimum thickness of the cell for concentration measurements seems to be 3 cm, from this figure.

The effect of the sample thickness on the isotopic determination was also studied. The results, shown in Table V, indicate that there is negligible difference with sample thickness; this shows that the MGA2 program is independent of sample thickness.

We have also tested the MGA2 code on plutonium solutions of different isotopic distributions to determine the accuracy of MGA2. Samples C and D were tested using the 3-cm-thickness cell. The results are shown in Table VI. For a measurement of 3600 s, the MGA2 method provides better than 1% accuracy for every isotope of plutonium.

We should point out that the MGA2 code determines the ^{242}Pu content by means of isotopic correlation from the other isotopes and is dependent on the reactor type from which the plutonium is produced.³ If the reactor type is known, then this information can be part of the input which should improve the agreement on this isotope, which should also improve the agreement on the other isotopes.

Fig. 3. The measured ratio of the 111- and 148-keV peak areas as a function of the sample thickness.

Table V. The effect of the sample thickness for the MGA2					
	Mass (wt%)	1 cm	2 cm	3 cm	5 cm
^{238}Pu	1.4	1.5	1.5	1.5	1.6
^{239}Pu	57.0	59.2	59.0	57.6	58.1
^{240}Pu	26.9	25.7	26.0	28.0	25.6
^{241}Pu	9.3	9.4	9.3	8.5	10.1
^{242}Pu	5.4	4.2	4.2	4.4	4.7

Table VI. Comparison between MGA2 and mass spectrometry determination of plutonium concentration				
	Sample C (wt%)		Sample D (wt%)	
	Mass Spec.	MGA2	Mass Spec.	MGA2
²³⁸ Pu	1.4	1.5 ± 0.01	0.8	0.9 ± 0.08
²³⁹ Pu	57.0	57.6 ± 0.75	66.0	67.1 ± 0.71
²⁴⁰ Pu	26.9	28.0 ± 0.16	22.1	20.2 ± 0.60
²⁴¹ Pu	9.3	8.5 ± 0.04	8.1	9.3 ± 0.05
²⁴² Pu	5.4	4.4 ± 0.1	3.0	2.5 ± 0.1

V. EXPERIMENTAL SETUP—PART 2

In the second part of the experiment, we investigated the application of the ID technique to measurement of plutonium concentration in stainless steel pipes. This continues the evaluation of the in-plant analytical technique.

The experimental setup with the pipe is shown in Fig. 4 below. In this experiment, the pipe is 29.9 mm in diameter, has a 2.3-mm-thick wall, and is 50 mm high. The size of the pipe is the same as that used at the main plant at which this in-plant equipment could be installed. The collimator hole diameter is 5 mm and its length is 10 mm. The front of the collimator and the inside of the hole were lined with 1-mm-thick cadmium. The assay time was 3600 s.

Fig. 4. Experimental setup for the measurement with pipes. The pipes are contained in double plastic bags.

A. Plutonium Concentration Versus Peak Ratio

The concentrations of plutonium used in this part of the experiment are 128, 149, and 174 g/l. The plutonium concentrations versus the 111/148 peak ratios are shown in Fig. 5. The variation of the ratio is less than that of the previous experiment because in a cylindrical pipe, part of the plutonium solution near the edge of the pipe will undergo little attenuation and thus does not contribute to the concentration determination.

We have also repeated one of the samples with another detector, which is equipped for high-rate counting. The following data, shown in Table VII, were collected for 3600 s at 9500 cps.

For the high-counting-rate measurement, the precision of the peak ratio from repeated runs is 0.34%. The precision of the plutonium concentration determination is, therefore, 1.7%. The count rate can be increased by a factor of 2, reducing the precision to 1.2% for a 1-h measurement.

Fig. 5. Plutonium concentration as a function of the measured 111/148 peak ratios. Plutonium solutions are in stainless steel pipes.

TABLE VII. Peak areas and ratios with a high-count-rate detector measuring at an input rate of 9500 cps.

Run Number	111 keV Area	148 keV Area	111/148 Peak Ratio
1	816920	601960	1.357
2	811487	600780	1.351
3	809363	600405	1.348
4	811623	603248	1.345
5	808622	602851	1.341
6	812033	600751	1.352
7	810725	603277	1.344
8	812032	602998	1.347
9	811565	602737	1.346
10	814141	602413	1.351
Average	811851	602142	1.348
St. Dev.	2337	1106	0.005
RSD%	0.29	0.18	0.34

B. Intrinsic Densitometry with Aged Plutonium

We have tested this technique with aged plutonium solution. Freshly separated plutonium with ~10% ^{240}Pu was allowed to decay for three weeks for americium buildup. Because of the relatively intense 60-keV peak from ^{241}Am , a 1-mm-thick cadmium absorber was used to reduce the counting rate from this peak. The plutonium solution concentration was 210 g/l. We want the same dimension stainless steel pipe as in the previous pipe measurement. The data are summarized in Table VIII.

The measurement precision was found to be worse than for the freshly separated plutonium. The reason is that the cadmium absorber that reduced the 60-keV count rate also reduced the counting rate at 111 keV and at 148 keV, but less than at 111 keV. The method can be used to determine the plutonium concentration of aged plutonium but with reduced precision.

VI. CONCLUSIONS

We have evaluated the potential application of the ID technique in a plant environment for process monitoring. We found that the ID method can determine the plutonium concentration to 1-2% from 100 g/l to 200 g/l with quartz cells and a measurement time of 3600 s. We also found that nitric acid concentration and the impurity level of uranium in the product plutonium solution does not affect the concentration measurement.

When this technique is applied to plutonium solutions in stainless steel pipes, we found that similar precision in plutonium concentration can be achieved using a high-count-rate detector. The precision, however, is reduced with aged plutonium solutions.

Table VIII. Peak areas and ratios from the aged plutonium solution			
Run Number	111-keV Area	148-keV Area	111/148 Peak Ratio
1	24564	44395	0.553
2	30790	50527	0.609
3	29849	50464	0.591
4	30501	50948	0.599
5	29852	50531	0.591
6	29878	51153	0.584
7	30734	51809	0.593
8	30921	51050	0.606
9	31486	51494	0.611
10	30407	51163	0.594
Average	29898.2	51015	0.593
St. Dev.	1948	459	0.017

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