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INTRINSIC DENSITOMETRY TECHNIQUES FOR HIGH-BURNUP PLUTONIUM SOLUTIONS

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ABSTRACT

Two intrinsic densitometry techniques for determining plutonium concentrations (and isotopic distributions) that require no external radioactive sources or x-ray generators and rely only on the natural radiation from the plutonium have been tested with high-burnup solutions. We found that these techniques can determine the plutonium concentration of high-burnup solutions to between 2.4% and 3.5% in a 1-h assay.

INTRODUCTION

An accurate measurement of the plutonium concentration of a sample is always necessary for nuclear material control and accounting. Our previous paper¹ discusses two methods of determining the low-burnup plutonium concentration (and isotopic distributions) that require *no* external radioactive sources or x-ray generators, but rely only on natural radiation. The methods are ideally suited to assay reasonably pure plutonium solutions, such as the product solutions of reprocessing plants and the eluate solutions from anion exchange columns. The methods can be applied to aged or freshly separated plutonium and can be used to measure plutonium concentrations in pipes or tanks.

The purpose of this work is to extend the methods of determining the plutonium concentration to high-burnup solutions. Both methods have been tested: the densitometry method and the ratio method, and in this paper the experimental results of these two methods are discussed. Finally, the potential application of these techniques is described.

EXPERIMENTAL SETUP

A set of six high-burnup solutions was prepared from the same plutonium isotopic batch, with concentrations ranging from 50 g/L to 310 g/L. The solution was pipetted into a spectrometric cell, which was placed in a secondary container as shown in Fig. 1. The inner diameter of the spectrometric cell was 2 cm and it was 2 cm long, holding approximately 6 mL of solution. The container was placed in a glove box and viewed by the detector, placed on the axis of the cell, through the glove box wall (plastic wall thickness 0.7 cm); the distance from the detector surface to the solution surface was approximately 7 cm. A seventh solution was prepared from a substantially different isotopic batch.

Each of the solutions was assayed for 3600 s for 7 to 16 cycles; some of the solutions were measured again later to test reproducibility. The solution was viewed by a side-looking



Fig. 1. Sample cells used for the measurement.

low-energy photon spectrometer detector with a 2-cm³ active volume and a resolution of 500 eV at 122 keV. The plutonium concentrations were characterized by the titration technique; the isotopic distribution was characterized by mass spectrometry. The concentrations were also confirmed by the K-edge densitometer designed for the product solutions.³ The concentrations of these solutions and sample thicknesses are listed in Table I. In this table, the first six samples all have the same isotopic distribution; the seventh has a different isotopic composition. They are listed in Table II. The counting rate ranges from 4000 to 15 000 counts/s.

Sample	Concentration (g/L)	Thickness (cm)
1	50.36	1.974
2	101.54	1.981
3	151.52	1.977
4	208.09	1.973
5	253.44	1.974
6	308.61	1.981
7	143.92	1.978

Sample	²³⁸ Pu (wt %)	²³⁹ Pu (wt %)	²⁴⁰ Pu (wt %)	²⁴¹ Pu (wt %)	²⁴² Pu (wt %)
1-6	2.002	54.541	25.257	11.771	6.429
7	0.228	70.078	24.249	4.398	1.13

The experimental work described in this paper was carried out at the European Institute for Transuranium Elements, Karlsruhe, Federal Republic of Germany.

FIRST METHOD--DENSITOMETRY

The first method of plutonium concentration determination without external sources uses the MGA2 isotopic program developed by R. Gunnink of Lawrence Livermore National Laboratory.² In MGA2, a relative efficiency curve is fitted with 10 peaks from 59 keV to 208 keV (59, 94, 101, 103, 110, 129, 148, 165, 203, and 208 keV), including the discontinuity at the plutonium K-absorption edge at 121.8 keV. Detailed discussion of the method can be found in Ref. 2. For a fixed solution sample thickness, the magnitude of the discontinuity should be proportional to the plutonium concentration.

The results are summarized in Table III. We found that the discontinuity is proportional to the plutonium concentration for a fixed sample thickness. Excluding the sample with a concentration of 50 g/L, the plutonium concentration can be determined by this method to 3.5% with a precision between 3% and 17% in a 3600-s assay. The average ratio of densitometry to chemistry (0.81) is consistent with the ratio of 0.79 determined in our previous work on low-burnup solutions. The error quoted in this work is the standard deviation of the repeated runs.

Chemistry (g/L)	Densitometry (g/L)	Densit/Chem	Error (%)
50.36	26.2	0.52 ^a	34
101.54	80.7	0.80	17
151.52	128.12	0.85	4.6
208.09	163.8	0.79	4.7
253.44	205.2	0.81	2.6
308.61	237.92	0.77	7.9
145.92	121.43	0.83	4.8
Av		0.81	
RSD		3.5%	

^aExcluded.

SECOND METHOD--RATIO TECHNIQUE

The second method uses the ratio of a pair of gamma- or x-ray peaks, one above the K-absorption edge and one below

the edge, so that the absorption coefficients are substantially different. For high-burnup solutions, the 129-keV peak is relatively weak; in comparison, the 148-keV peak from ²⁴¹Pu is much more intense. The μ values for the plutonium 148-keV gamma (²⁴¹Pu) and the 111-keV x-ray (UK β_1 from ²³⁸Pu, ²³⁹Pu, and ²⁴¹Pu) differ by 1.29 cm²/g. Because of the difference in the μ values, the ratio of the 111/148 peak intensities is a function of the plutonium concentration. For a fixed-solution thickness, this function can be used to determine the plutonium concentration. These two peaks are selected for high-burnup plutonium because they bracket the plutonium K-absorption edge and are only 37 keV apart. A typical plutonium spectrum in this energy range is shown in Fig. 2. Observe that the UK β_1 and UK β_3 peaks are well separated from the other peaks, and the sum of these two peaks is used for the 111-keV x-ray peak.

To test this method, the 111-keV net peak area and the 148-keV net peak area from the previous experimental data

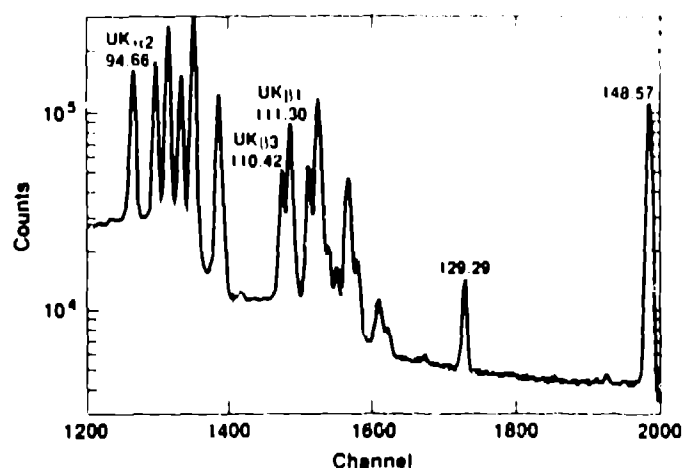


Fig. 2. Typical spectrum from low-burnup plutonium solution in the 94- to 148-keV region.

were also extracted with the MGA2 code. The data require an isotopic correction, which will be discussed below.

Mathematical Model

To understand the behavior of the ratio technique, we developed a simple model. In this model, the detector is assumed to be far away from the solution sample as shown in Fig. 3.

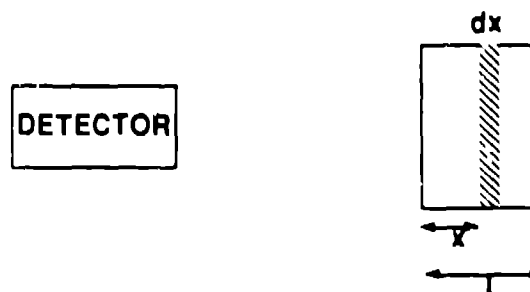


Fig. 3. Simple mathematical model of the measurement.

TABLE IV. Plutonium concentration determined from the 111/148 ratio for 2-cm-thick solutions						
Chemistry (g/L)	No. of runs	Ratio (111/148)	Error in ratio (%)	Pu Conc. from ratio (g/L)	Error in conc. (%)	Pu Conc. Chem.
50.36	16	0.89614	0.53	52.04	9.3	1.04
101.54	16	0.93617	0.20	96.91	1.9	0.95
151.52	7	0.98434	0.41	154.98	2.8	1.02
151.52	4	0.98236	0.53	152.50	3.6	1.01
208.09	7	1.02606	0.28	208.87	1.5	1.00
208.09	3	1.02449	0.24	206.79	1.3	0.99
253.44	16	1.05791	0.24	252.27	1.2	1.00
308.61	7	1.09917	0.23	311.39	1.0	1.01
308.61	2	1.09552	0.32	306.03	1.4	0.99
145.92	16	0.97349	0.70	141.51	4.8	0.97
Av						1.00
RSD						2.4%

the ratio technique, which is defined as the fractional change in plutonium concentration per fractional change in the ratio of 111/148, is a function of both the plutonium concentration and sample thickness. The sensitivity is shown in Fig. 5. The error on the plutonium concentration determination is the product of the error of the ratio and the sensitivity for the 2-cm-thick sample. The error or precision can be improved by a higher counting rate, which can be increased by a factor of 2.

We found that the plutonium concentration of high-burnup solutions can be determined with a relative standard deviation of 2.4% from 50 g/L to 310 g/L by the ratio technique.

CONCLUSIONS AND APPLICATIONS

The main advantage of the two techniques described is that no external radioactive source or x-ray generator is required. The experimental data in this work demonstrate that for high-burnup plutonium, the concentration can be determined reasonably well in a 1-h assay. However, these techniques are not for all plutonium solutions; if the solution is reasonably pure, the techniques can be used to determine the plutonium concentration. The product solution of a reprocessing plant and the product solution from an anion exchange are ideal candidates for these techniques.

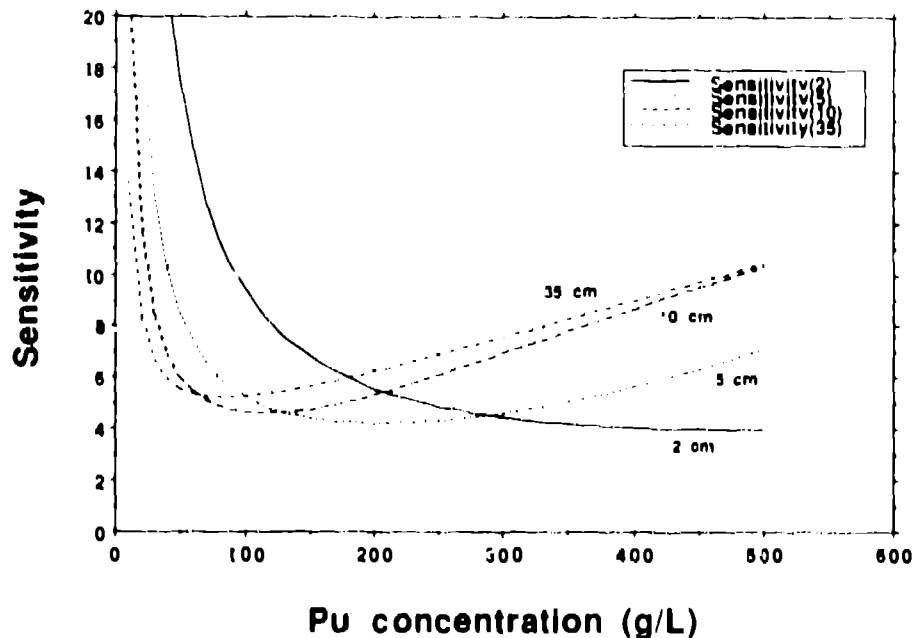


Fig. 5. Sensitivity as a function of plutonium concentration and sample thickness.

Because the techniques rely only on the natural radiation from plutonium, the assay setup can be extremely simple, and the techniques have several important applications.

They can be used by inspectors to verify the reprocessing plant product solutions (both the isotopes and the concentration). The fact that the methods require no external source is a tremendous operational convenience. We recommend the following inspection sequence:

- The inspector should establish the K^* calibration constant and the quadratic curve for the ratio technique. This should be done with the detector designated for inspection, and can be done at the laboratory under the inspector's control. The calibration constant, K^* , depends on the detector.
- At the facility where the inspection is to be carried out, the facility personnel will use the cells with known thicknesses to hold the solutions to be verified.
- If there are differences in the absorbers between the calibration and the inspection, the effect on the calibration constant can be calculated.
- The plutonium concentrations can be determined by the $^{241}\text{Pu}/^{242}\text{Pu}$ measured ratios using the quadratic equation. The quadratic equation determined in this work can be used.

Another potential application of the ratio technique is to monitor the plutonium concentration in holding tanks.

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- ρ = density of plutonium in g/cm³
 μ_{111}, μ_{148} = mass absorption coefficients of plutonium
 μ_{111}^m, μ_{148}^m = mass absorption coefficients of the matrix
 ρ_m = density of the matrix
 $\epsilon_{111}, \epsilon_{148}$ = detection efficiencies
 f_8, f_9, f_0, f_1, f_2 = weight fractions of plutonium isotopes
 g_1 = γ /s-g of ²⁴¹Pu
 x_8, x_9, x_0, x_1, x_2 = x/s-g of (111 keV) of plutonium isotopes.
 A = cross-sectional area

Detection of 148-keV gamma ray:

$$\begin{aligned}
 I(148) &= \epsilon_{148} \rho g_1 f_1 A \int_0^L \exp[-(\mu_{148} \rho + \mu_{148}^m \rho_m)x] dx \\
 &= \epsilon_{148} \rho g_1 f_1 A \frac{L}{\mu_{148} \rho + \mu_{148}^m \rho_m} \\
 &\cdot \left\{ 1 - \exp \left[-(\mu_{148} \rho + \mu_{148}^m \rho_m)L \right] \right\} \\
 &= \epsilon_{148} \rho g_1 f_1 AL \frac{1 - T_{148}}{\ln(T_{148})}
 \end{aligned}$$

The above expression is correct for the far-field approximation. The more accurate expression is given below, where $CF(AT)$ is the correction factor for attenuation.

$$I(148) = \epsilon_{148} \rho g_1 f_1 AL \frac{1}{CF(AT)_{148}}$$

Similarly, for the detection of the 111-keV x-ray:

$$I(111) = \epsilon_{111} \rho L \left(\sum_i x_i f_i \right) A \frac{1}{CF(AT)_{111}}$$

The ratio 111:148 is

$$\frac{I(111)}{I(148)} = K \frac{\sum_i x_i f_i}{g_1 f_1} \frac{CF(AT)_{148}}{CF(AT)_{111}}$$

$$\text{where } K = \frac{\epsilon_{111}}{\epsilon_{148}},$$

$$= K^* \cdot \left(1 + \frac{f_8 x_8}{f_1 x_1} + \frac{f_9 x_9}{f_1 x_1} + \frac{f_0 x_0}{f_1 x_1} \right) \cdot \frac{CF(AT)_{148}}{CF(AT)_{111}},$$

$$\text{where } K^* = \frac{x_1 \epsilon_{111}}{g_1 \epsilon_{148}},$$

$$= K^* \cdot CF(ISO) \cdot \frac{CF(AT)_{148}}{CF(AT)_{111}},$$

where the $CF(ISO)$ is the isotopic correction factor.

The results are shown in Fig. 4. The plutonium concentration is plotted against the measured 111/148 ratio, corrected by dividing with the isotopic factor

$$1 + f_8/f_1 \cdot x_8/x_1 + f_9/f_1 \cdot x_9/x_1 + f_0/f_1 \cdot x_0/x_1,$$

where $x_8/x_1 = 0.1732$,
 $x_9/x_1 = 0.0084$, and
 $x_0/x_1 = 0.00136$.

The data fit a quadratic equation well, indicating that if the plutonium isotopic distribution is known or measured, the 111/148 ratio can be used to determine the plutonium concentration.

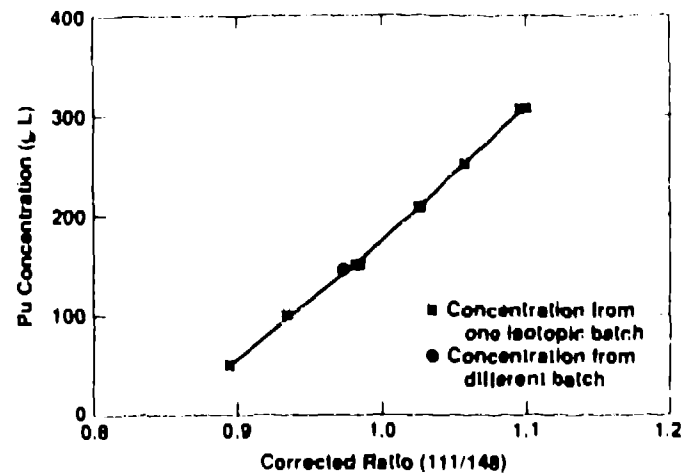


Fig. 4. The plutonium concentration as a function of the measured 111/148 ratios after the isotopic correction.

The measured ratios, after the isotopic correction, are also listed in column 3 of Table IV. Also listed (column 4) is the error of the ratio, which is calculated from the standard deviation of the repeated runs. The error of the plutonium concentration determination, however, is greater. The sensitivity of