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AN INSTRUMENT FOR MEASUREMENT OF Pu CONCENTRATION AND ISOTOPICS OF PRODUCT SOLUTIONS AT TOKAI-MURA

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Abstract

Rapid analysis of product solutions for plutonium concentrations and isotopic distributions is accomplished by a new instrument currently installed at the reprocessing plant in Tokai-mura, Japan. Better than half-percent measurements of the Pu concentration are obtained by K-edge densitometry for concentrations ∿250 g Pu/ℓ in short count times. Isotopic fractions are determined with an accuracy of one percent or better in an additional 30-minute passive count.

1. Introduction

A new instrument based on gamma-ray nondestructive analytical techniques is undergoing testing and evaluation for plutonium product solution measurements at the Tokai-mura reprocessing facility. The Los Alamos Scientific Laboratory (LASL)-designed instrument was developed for plant operations under the TASTEX program. Performance capabilities determined by preliminary testing at LASL have recently been confirmed by the product solution measurements at Tokai-mura. The testing and evaluation has been a collaborative effort of the Power Reactor and Nuclear Fuel Development Corporation (PNC) and LASL.

The instrument design is reviewed briefly. Preliminary calibration results from LASL and Tokai-mura are summarized and compared with results from the first reprocessing campaign.

2. Instrument Design

The Tokai-mura instrument operates off-line beneath the glove box to which process solution samples are pneumatically transported. K-edge densitometry determines the concentration of plutonium in solution. The instrument uses 121.1- and 122.1-keV gamma rays from ^{75}Se and ^{57}Co radioisotopic sources, respectively, to obtain two transmission measurements at these energies which bracket the plutonium K-edge. The sources with appropriate collimators are positioned sequentially for measurements in the transmission geometry by a motor-driven Geneva mechanism. Gamma radiation is detected and analyzed by a high-resolution spectrometer that uses a thin planar intrinsic germanium crystal. Except for sample positioning, the analysis procedure is entirely automated and under computer control.

The arrangement of equipment beneath the glove box is shown schematically in Fig. 1. The counting well is an extension from the base of the glove box. Solution samples in disposable vials are positioned in the well in the transmission path. The concept of the transmission measurement is shown in Fig. 2, which illustrates the basic mechanical components.

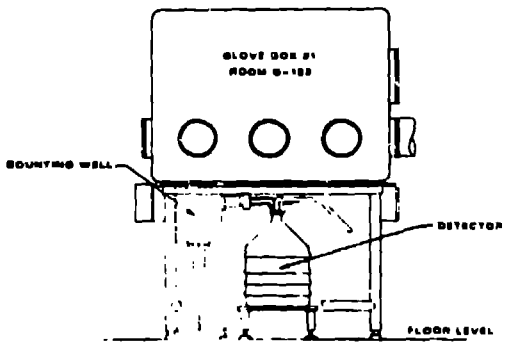

Fig 1. Schematic view of measurement station extending below and positioned beneath glove box.

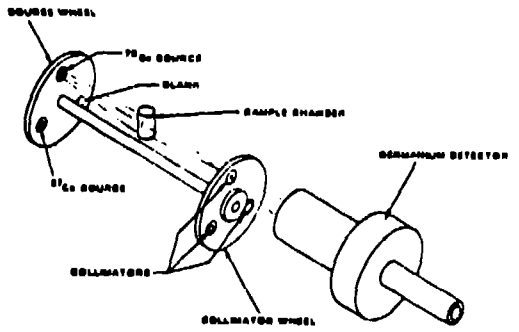

Fig. 2. Conceptual view of measurement configuration. The sample is shown between the source (^{75}Se, in this case) and the collimator in the detector line of sight. Sources and collimators are mounted on separate wheels driven synchronously.

An additional option in the measurement is a complete plutonium isotopics analysis. A passive count can be performed with wide-open sample-to-detector collimation using a mechanical positioning such that both sources are removed from the transmission geometry and shielded from the detector. (Refer to Fig. 2.) Self-attenuation effects on the passive gamma rays are important because of the relatively large sample thicknesses (∿0.5 g Pu/cm^2). Correction factors for self-attenuation are determined and applied to the measured peak areas at 129, 149, and 153 keV corresponding to ^{239}Pu, ^{241}Pu, and ^{238}Pu, respectively. The determination of the correction factors involves the interpolation of measured transmissions of several gamma rays from ^{75}Se and ^{57}Co to the three energies of the passive assay peaks. The interpolated transmissions are used to determine the correction factors by

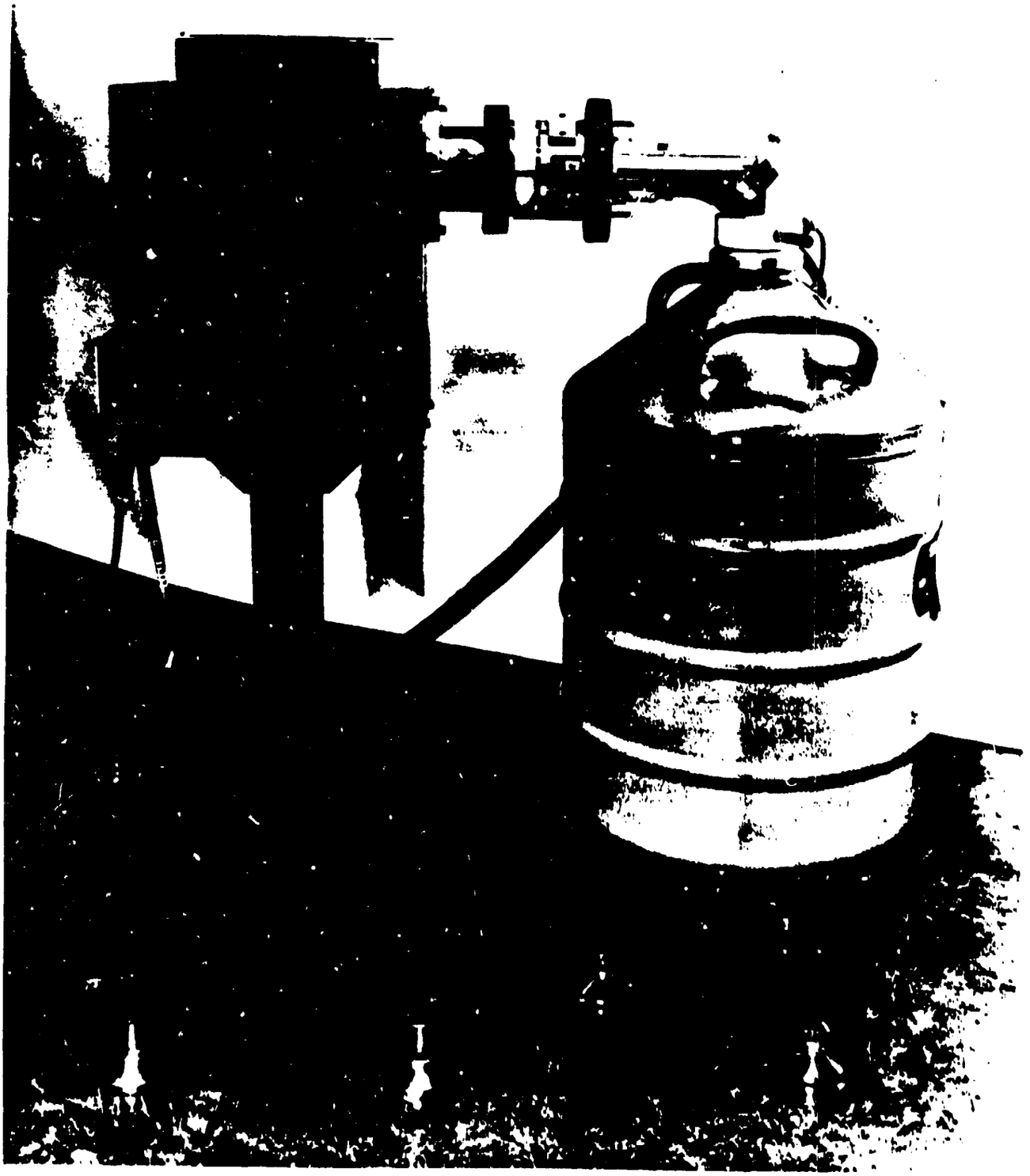

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Fig 3

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Fig 4

a technique demonstrated in existing instrumentation.[1] The low-energy region is used for analysis of ^{240}Pu and for a separate determination of ^{238}Pu. The assay gamma rays are at 43 and 45 keV, respectively. The self-attenuation effects are incorporated into relative efficiency measurements based on peak areas of the ^{239}Pu gamma-ray lines at 39 and 51 keV. The ^{242}Pu fraction, which cannot be determined by gamma-ray spectroscopy, is evaluated by isotopic correlation techniques. The isotopics analysis uses only ratios of closely-spaced peak areas so that the measurement is insensitive to counting geometry and absolute detection efficiency.

The equipment at the measurement station beneath the glove box is shown in a photograph reproduced in Fig. 3. The entire assembly of equipment for this instrument appears in Fig. 4.

Fig. 3. A close-up view of the measurement well and intrinsic germanium detector before installation at Tokai-mura.

Fig. 4. Complete K-edge densitometer including computer, multichannel analyzer CRT and NIM electronics (electronics rack at left), hardcopy terminal (center), and measurement station equipment (right), before installation at Tokai-mura.

The electronics rack (left) and hardcopy terminal are located in another portion of the room.

More details of the instrument design and measurement principles are published elsewhere.[2,3]

3. Results

Calibration of the Densitometer

Well-characterized plutonium solutions were used as reference materials for preliminary calibration of the densitometer at LASL. Plutonium concentrations varied between 150 and 300 g per liter. The ^{240}Pu isotopic content of the solutions was 15 percent although the calibration was also verified with solutions of low-burnup material. Figure 5 shows the results plotted for each measurement as the percent deviation from the mean calibration parameter. The one-half percent error bars showing the calculated statistical uncertainty ($\pm 1\sigma$) in each measurement are consistent with the scatter in the data.

The long-term stability of the densitometry calibration is demonstrated by repeated measurements of a secondary standard plutonium foil with a thickness equivalent to that of the solutions measured by the instrument. Figure 6

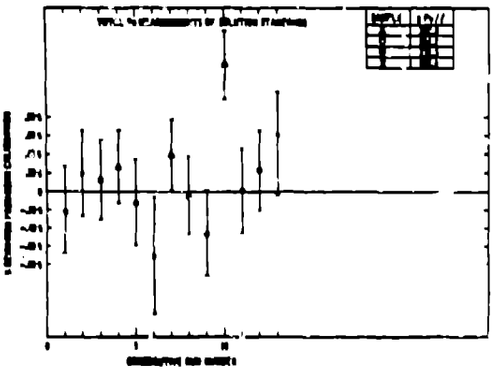

Fig. 5. Results of the densitometer calibration at LASL. Measurements were performed with well-characterized solutions of high-burnup plutonium.

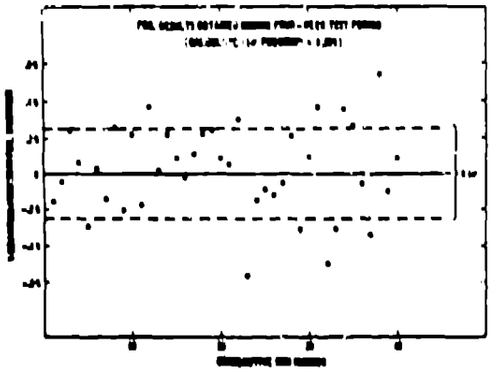

Fig. 6. Foil results determined by densitometry during a 4-week test period at LASL.

is a plot of the foil data obtained during a one-month period at LASL. The scatter in the foil assay agrees with the statistical prediction. No biases were observed to appear over a measurement period of several months. The foil assay is a daily measurement control requirement for routine operation of the densitometer.

The densitometer was calibrated at Tokai-mura in October 1979 just after installation. The calibration results are shown in Fig. 7. Plotted versus plutonium concentration are the percentage differences between the densitometry measurement and the analytical chemistry reference value used to establish the calibration. The error bars reflect the statistical precision ($\pm 1\sigma$) in the densitometry assay. The analytical chemistry precision is approximately plus or minus one-half percent. The observed scatter of this magnitude is consistent with the combined precisions. The densitometer data were obtained in 1500-s count times.

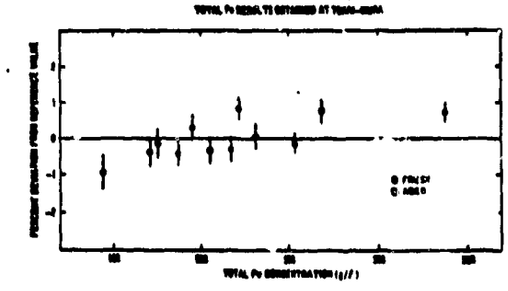

Fig. 7. Results of densitometer measurements of reference solutions at Tokai-mura. "Fresh" and "aged" correspond to two weeks and one year, respectively, after americium separation. The precision (1σ) in these data is approximately one-half percent.

TABLE I
PRECISIONS IN ISOTOPIC FRACTIONS
FOR HIGH-BURNUP (15% ^{240}Pu) REFERENCE SAMPLES

ppm americium	100	100	1000	3000
Pu Isotope	1σ (calc.) / 1σ (expt.)	Calculated Precision (%, 1σ)		
238	1.2	0.9	1.2	1.3
239	1.0	0.45	0.45	0.45
240	1.2	1.0	1.3	1.5
241	0.7	0.9	0.9	0.9
242	*	*	*	*

*^{242}Pu fraction is derived from a correlation based upon archival isotopics data. The precision derived from the data base is ± 40 percent.

Isotopics Calibration

Table I shows the results of the isotopics calibration at LASL using the 15 percent ^{240}Pu solutions. Reference values were the mass spectroscopic results. Column 2 compares the average calculated measurement precision with the experimental scatter in the weight fractions for each isotope. The results are close to unity indicating no significant contributions to the measurement precision beyond the statistical contributions. The measurements were performed within several days of the americium separation so that the ^{241}Am content was about 100 ppm.

As the solutions age and ^{241}Am grows in, the statistical uncertainty in the results for the even isotopes increases. Data obtained with the same solutions after aging are shown in Table I, columns 4 and 5, the latter corresponding to a 2-month aging of the high-burnup material. Since the ^{241}Pu content of spent fuel varies with burnup, the effect of ^{241}Am at a given aging time will vary with burnup. Passive measurements of 15 percent ^{240}Pu material are feasible for aging times up to about 6 months.

The calibration for isotopics is primarily a measurement of relative detection efficiencies of gamma rays closely spaced in energy. The measurement control procedure which verifies the long-term stability of the isotopics calibration is the determination of the relative efficiency of the 121- and 136-keV gamma rays from the ^{75}Se source in the absence of a sample. This measurement is performed daily during the no-sample counting of the transmission sources.

A verification of the isotopics calibration was performed at Tokai-mura in October 1979 using a single mass spectroscopic determination as a reference value. The precisions for the isotopics analysis determined at this time were in agreement with those measured at LASL.

Process Solution Measurements

Densitometry results were obtained from measurements of seven process solution samples during the campaign in November and December 1979. The results were compared with analytical chemistry measurements of these samples. The precision in the percentage differences between densitometry and chemistry is one-half percent (1σ), consistent with the combined precision for these densitometry and chemistry determinations. A positive bias of one-half percent in the average difference indicates a shift in vertical positioning of the disposable (tapered width) sample cells. The shift is probably caused by a change in vertical position of the plastic liner in the well. The liner is normally clamped rigidly in place. The daily measurement of the calibrated plutonium foil will monitor any future change in position. However, the foil was not available at Tokai-mura until January 1980.

Isotopics data were obtained with five freshly separated process solutions covering a range of high burnup. The five mass spectroscopic determinations were used to establish

accurate calibration parameters. The results with the new parameters are shown for each isotope in Table II. The precision is determined by repeated measurements of each sample. The accuracy for ^{238}Pu is not shown because of a possible mass 238 contaminant in the mass spectroscopic measurement.

TABLE II
SUMMARY OF ISOTOPICS RESULTS
FOR FIVE PRODUCT SOLUTION SAMPLES

Pu Isotope	Average Deviation (%) from Mass Spectrometric Result[a]	Precision (%, 1σ)
238	---	±0.9
239	-0.08 ± 0.21	±0.2
240	-0.14 ± 0.51	±0.7
241	0.04 ± 0.93	±0.6
242	7. ± 13.	±2.0

[a]Error is the scatter (1σ) in the percent deviation determined for each of the five samples.

4. Conclusions

Preliminary data demonstrate the precisions that can be achieved in rapid, nondestructive assays of product solutions for total plutonium and isotopics. Continued evaluation is necessary to verify the long-term stability of the calibration. The in-line performance of an instrument of similar design is soon to be evaluated at the Savannah River Plant.[4] The combined results of these in-plant evaluations should provide new information on safeguards capabilities for NDA in reprocessing.

5. References

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4. H. A. Smith, Jr., T. Marks, L. Cowder, C. Shonrock, S. Johnson, R. Slice, J. Sprinkle, K. W. MacMurdo, R. L. Pollard, and L. B. Baker, "Development of In-Line Plutonium Solution NDA Instrumentation at the Savannah River Plant (SRP)," Proc. 2nd Annual ESARDA Symp. on Safeguards and Nuclear Material Management, Edinburgh, Scotland, March 26-28, 1980 (to be published).

Figure Captions

1. Schematic view of measurement station extending below and positioned beneath glove box.

2. Conceptual view of measurement configuration. The sample is shown between the source (^{75}Se, in this case) and the collimator in the detector line of sight. Sources and collimators are mounted on separate wheels driven synchronously.

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