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In-Plant Measurements of Gamma-Ray Transmissions for Precise K-Edge and Passive Assay of Plutonium Concentration and Isotopic Fractions in Product Solutions

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CONTENTS

IN-PLANT MEASUREMENTS OF GAMMA-RAY TRANSMISSIONS FOR PRECISE K-EDGE AND PASSIVE ASSAY OF PLUTONIUM CONCENTRATION AND ISOTOPIC FRACTIONS IN PRODUCT SOLUTIONS

(FINAL REPORT ON TASTEX TASK G)*

by

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ABSTRACT

An instrument based upon high-resolution gamma-ray measurements has been tested for more than 1 year at the Tokai Reprocessing Facility for determination of plutonium concentration by K-edge absorption densitometry and for determination of plutonium isotopic fractions by transmission-corrected passive gamma-ray spectrometry. The nondestructive assay instrument was designed and built at Los Alamos National Laboratory for the Tokai Advanced Safeguards Technology Exercise (TASTEX). It was used at Tokai for the timely assay of more than 100 product solution samples during the TASTEX evaluations. The results were compared to reference values obtained by conventional destructive analysis of these samples. The precision and accuracy of plutonium concentrations measured by the K-edge technique are shown to be within 0.6% (1σ) in these applications. The precisions and accuracies of the isotopic fractions determined by these passive gamma-ray methods are shown to be within 0.4% for ^{239}Pu, 1% for ^{240}Pu and ^{241}Pu, and 10% for ^{242}Pu.

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*This publication does not necessarily reflect the views of the TASTEX Steering Committee.

I. INTRODUCTION

A. Purpose

One of the most ambitious technical ventures in international safeguards cooperation is the Tokai Advanced Safeguards Technology Exercise (TASTEX) for evaluation of plutonium accounting capabilities for safeguards at fuel reprocessing plants. Thirteen evaluation tasks were monitored by the TASTEX Steering Committee, which consists of representatives of Japan, the United States, France, and the International Atomic Energy Agency. The TASTEX Executive Summary Report[1] gives a detailed description of the TASTEX program and the individual tasks.

The Tokai Reprocessing Facility of the Power Reactor and Nuclear Fuel Development Corporation (PNC) in Tokai-Mura, Japan, was made available for testing and evaluation of certain TASTEX instruments, with full cooperation of PNC personnel in maintenance and operation of equipment, analytical support, and acquisition and evaluation of data. The evaluation of one such instrument (Task G of the TASTEX Program) is the subject of this report.

B. Methods

High-resolution gamma-ray spectrometry applied to measurement of transmissions of discrete gamma rays through samples of nuclear material is the basis for several techniques developed at Los Alamos National Laboratory for the nondestructive determination of concentration and isotopic content of plutonium in solution. One of these techniques is energy-dispersive K-edge absorption densitometry using radioisotopic transmission sources. Laboratory measurements of the ratio of transmissions of the discrete gamma rays of ^{75}Se (at 121.1 keV) and ^{57}Co (at 122.1 keV), on opposite sides of the plutonium K absorption edge (at 121.8 keV), were shown[2-4] to give precise and accurate assays of plutonium concentration in solution samples covering a wide range of plutonium concentration. The TASTEX demonstrations at Tokai included the first applications of this K-edge assay technique to process solutions, both aged and newly processed.

Measured transmissions of discrete gamma rays have been used at Los Alamos with considerable success[5] in providing self-attenuation corrections in the isotopics assay of an extremely wide range of plutonium solution concentrations. Application of transmission-corrected passive gamma-ray spectrometry to the complete isotopics determination of plutonium in newly processed solutions was demonstrated for the first time during the TASTEX evaluation at Tokai.

A nondestructive assay instrument based upon high-resolution gamma-ray measurements was tested off-line for more than 1 year at Tokai for determination of plutonium concentration by K-edge absorption densitometry and for determination of plutonium isotopic fractions by transmission-corrected passive gamma-ray spectrometry. The instrument was designed and built at Los Alamos under TASTEX. It was used at Tokai for the assay of more than 100 product solution samples during the TASTEX evaluation. The results were compared to reference values obtained by conventional destructive analysis of these samples.

C. Scope

This report reviews the design of the Tokai instrument and the procedures used for its evaluation under TASTEX. The measurement results are presented in detail, and the accuracies and precisions of the nondestructive assay techniques are determined for the Tokai applications in the context of the evaluation. Possible applications of this instrument for safeguards and materials accounting are discussed.

II. INSTRUMENT DESIGN

A. Equipment

The Tokai instrument was designed for off-line assay of solution samples for plutonium concentration and isotopics. The instrument is located in an analytical laboratory in the PNC plant at Tokai. The equipment consists of a computer-controlled multichannel analyzer (MCA); electronics and a hardcopy terminal, located in one corner of the laboratory; and measurement station equipment, located beneath a glovebox approximately 2.5 m from the electronics.

Figure 1 is a schematic drawing of the glovebox and measurement station. The glovebox is modified to include a well that extends downward from the glovebox floor. Solution samples in doubly contained vials are introduced into the glovebox through pneumatic transport tubes following removal of the solutions from the process. The solutions are transferred into disposable plastic measurement vials that are inserted into the well for assay. The operator uses the glovebox ports for all sample manipulations.

The measurement station consists of an intrinsic planar germanium detector (200 mm^2 by 7 mm) and a mechanical system. Figure 2 is a conceptual view of the mechanical system and detector. The ^{75}Se and ^{57}Co transmission sources, mounted in separate positions on a wheel, are rotated into the measurement position by a motor-driven Geneva mechanism. Collimators positioned between the sample and the detector are rotated synchronously into the transmission path. For the passive measurement, the sources are rotated out of the measurement position and shielded from the detector, and the collimation is enlarged. Figure 3 is a detailed drawing of the measurement well and mechanical system. The entire mechanical system is automated and under computer control.

A ^{109}Cd source, mounted on the detector in fixed geometry with respect to the crystal, is used to correct for (variable) losses of events resulting from changing count rates. The 88.0-keV gamma ray is monitored for this purpose.

The signals from the germanium detector preamplifier are shaped, amplified, and digitized by NIM modules in the electronics rack. Pileup pulses are electronically eliminated from the digitized spectrum. A two-point digital hardware stabilizer, under computer control, maintains a constant energy calibration. The microcomputer-based MCA stores the spectra and transmits the integrated counts in preset regions of interest to the computer for analysis following acquisition of each spectrum.

Figures 4 and 5 are photographs of the measurement station and the MCA, electronics, and hardcopy terminal in their respective positions at the PNC plant. Once a sample is positioned in the glovebox well, the operator specifies count times at the hardcopy terminal. The assays (K-edge and passive) then proceed automatically. The assay results (plutonium concentration and isotopic fractions) are printed at the hardcopy terminal. Spectra are stored automatically on the floppy diskette.

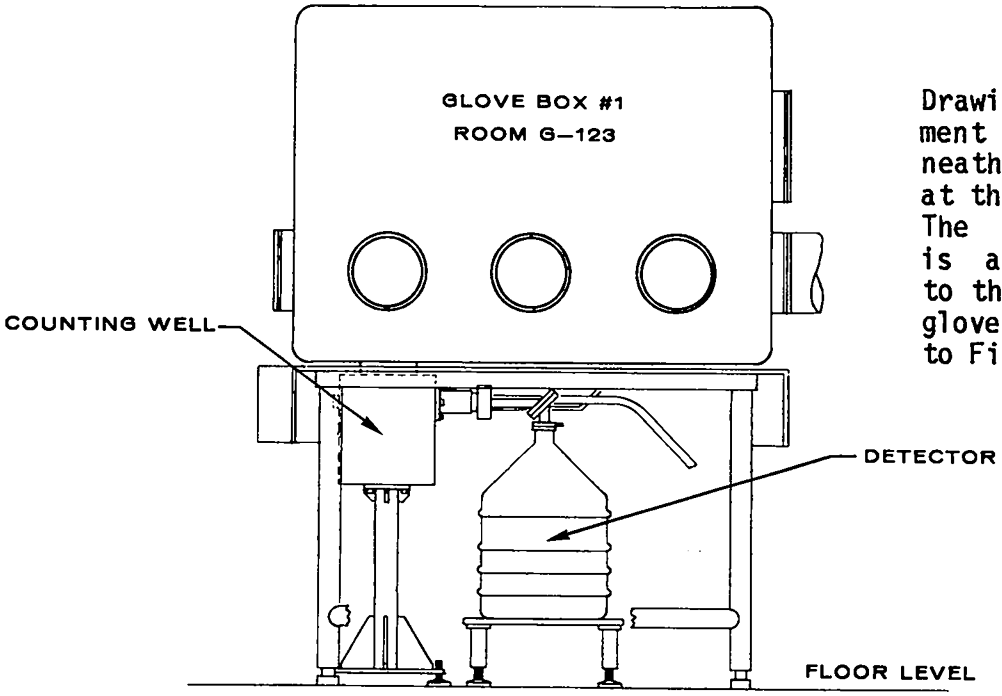

Fig. 1.
Drawing of measurement station beneath the glovebox at the Tokai plant. The counting well is a modification to the floor of the glovebox. (Refer to Fig. 3.)

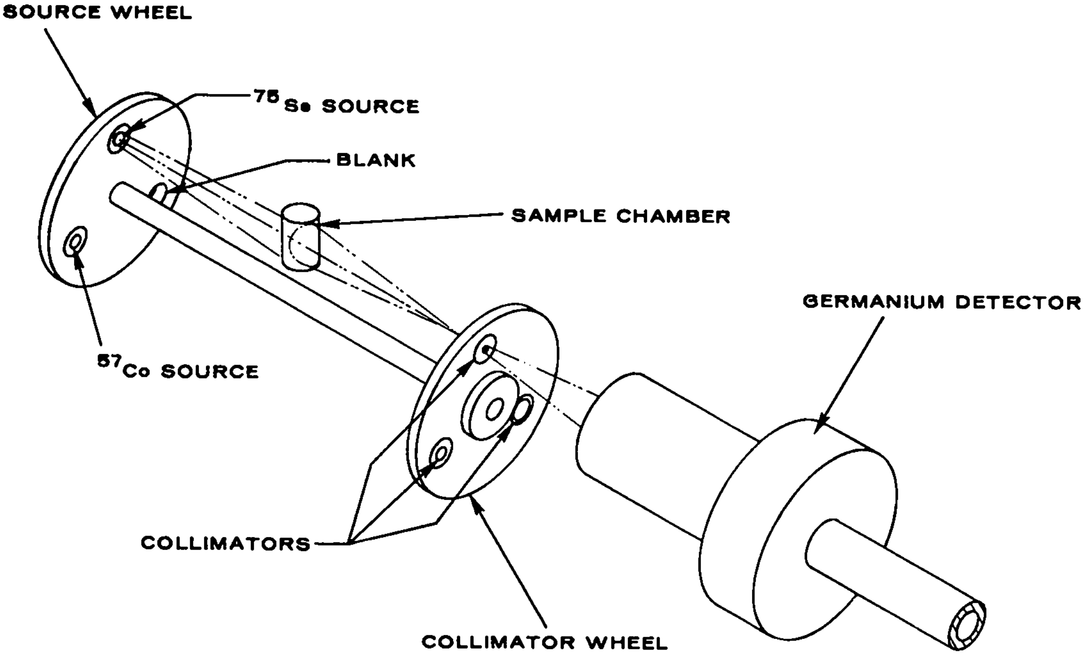

Fig. 2.
Illustration of the concepts of K-edge (transmission) and passive measurements. The sample chamber is shown in position (in the counting well) for measurement of the transmissions of the ^{75}Se gamma rays.

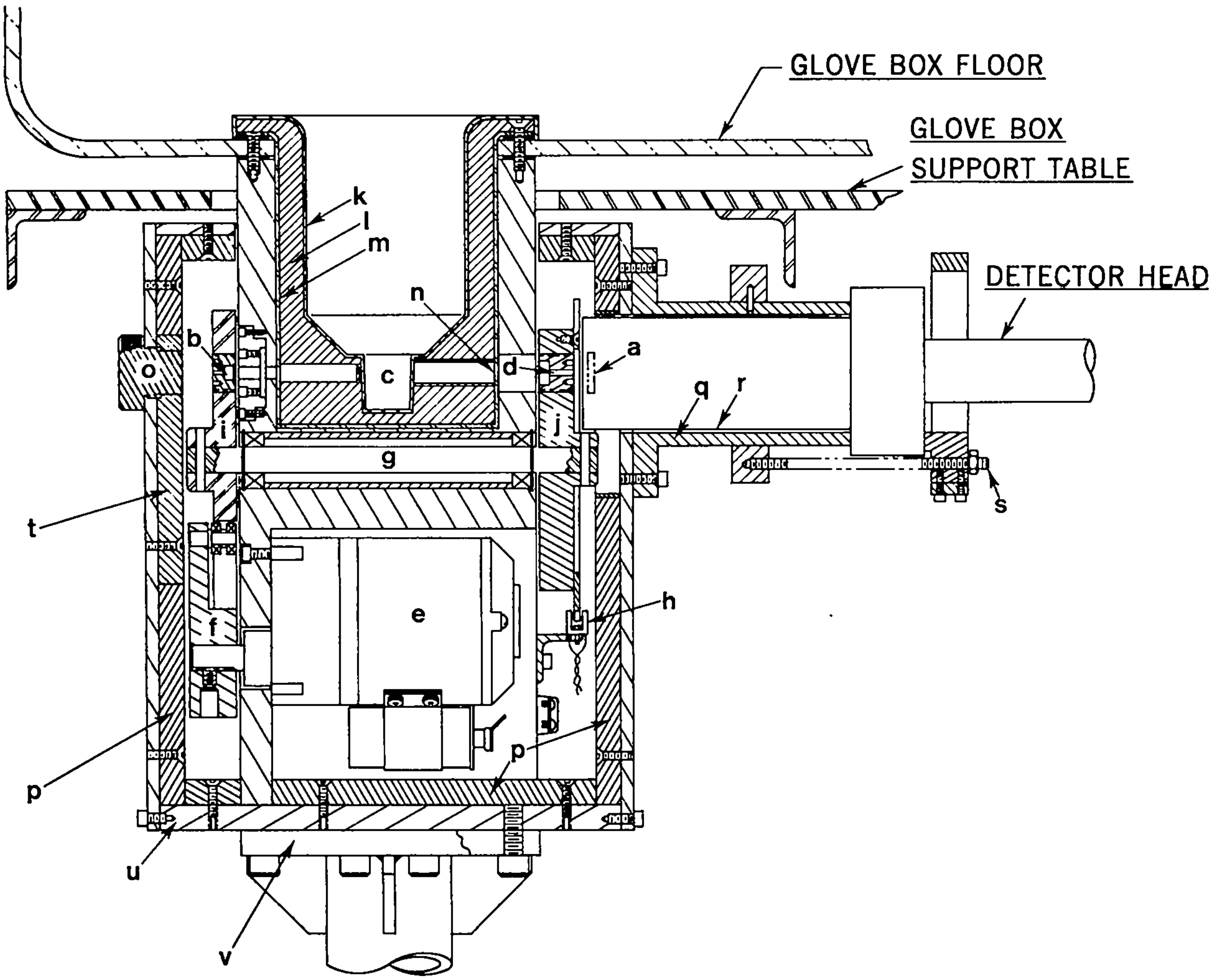

Fig. 3.
Detailed features of the mechanical system:

- (a) germanium crystal
- (b) tungsten transmission source holder
- (c) sample position
- (d) tungsten collimator holder
- (e) drive motor for Geneva mechanism
- (f) Geneva mechanism
- (g) common axle for source and collimator wheels
- (h) position-sensing mechanism
- (i) stainless steel source wheel
- (j) tungsten collimator wheel
- (k) polyurethane liner for measurement well
- (l) tungsten shield
- (m) stainless steel containment can
- (n) 0.25-mm-thick stainless steel window
- (o) removable tungsten plug for changing sources
- (p) lead shielding
- (q) tungsten detector shield
- (r) polyethylene electrical shield
- (s) detector clamping device
- (t) tungsten shielding
- (u) aluminum anchor plate
- (v) aluminum support base

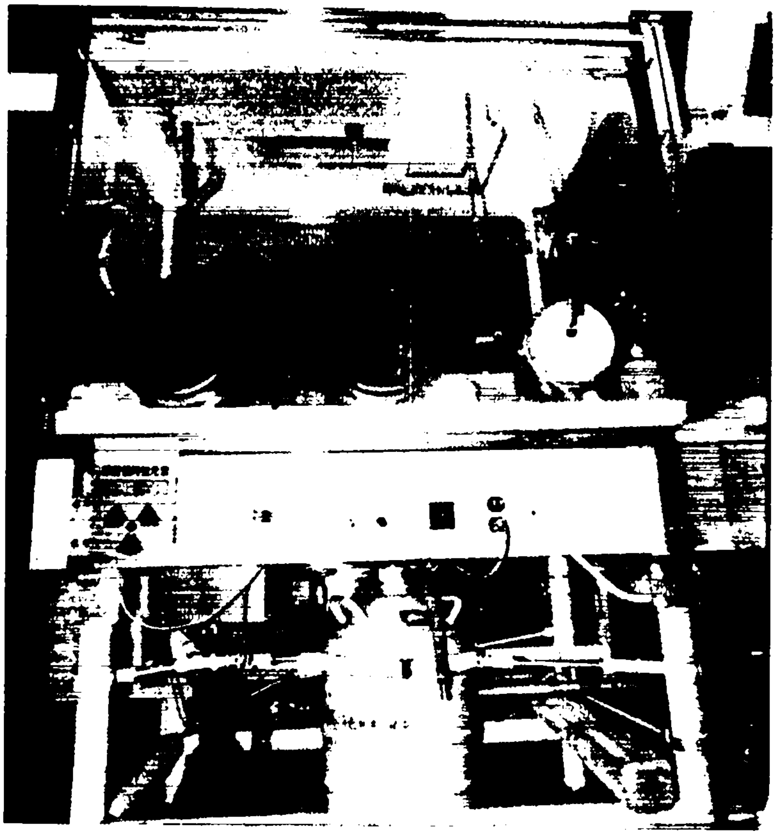

Fig. 4.
Photograph of the measurement station beneath the glovebox at the Tokai plant.

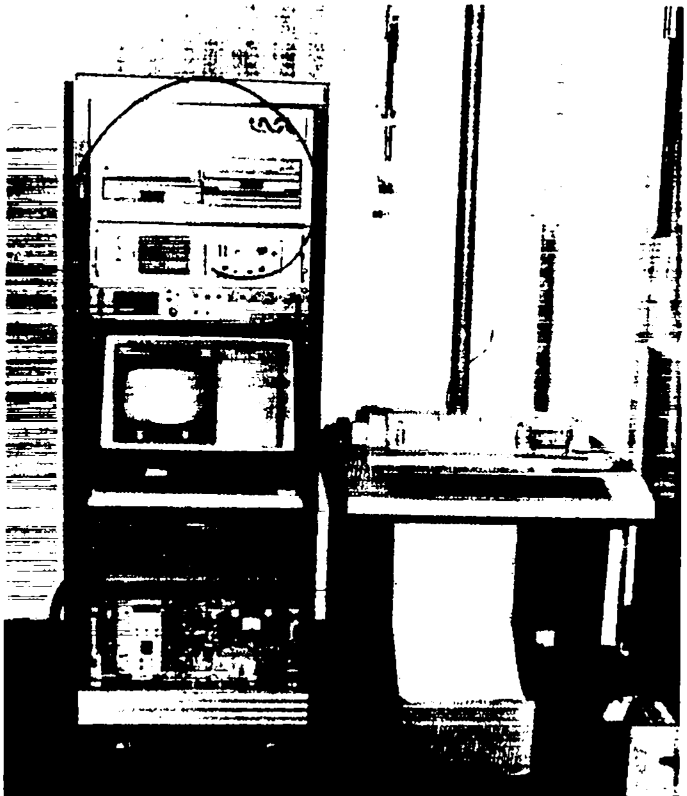

Fig. 5.
Photograph of the electronics and the hardcopy terminal at the Tokai plant. The electronics rack includes (top to bottom) a dual floppy disk drive, an oscilloscope, the control unit for the mechanical system, the multichannel analyzer with CRT terminal, the minicomputer, and NIM modular electronics.

Previous publications[6,7] have discussed the design of this equipment in greater detail.

B. Assay Sequence

1. Introduction. Routine operation of the Tokai instrument for assay of solution samples occurs in two programmed sequences. The first of these is measurement control, exercised once daily. The second is the measurement of spectra with solution samples positioned in the measurement well.

For each of these sequences, net peak areas are determined by subtraction of straight-line backgrounds obtained from two regions of interest that bracket the peak region. The peak region is, typically, three times the peak full width at half maximum (FWHM). The background regions are, typically, one-third the peak region width. The gamma-ray peaks for which regions of interest are set are listed in Table I. The amplifier gain is set so that 4000 channels correspond to approximately 300 keV. A formal description of the methods used for peak area determination is given elsewhere.[8]

The analog-to-digital converter is digitally stabilized by two-point monitoring of each of the spectra obtained in the sequences described below. The low-energy point applies to stabilization of the zero offset; the ^{109}Cd 88-keV peak is used because it appears in all spectra. The gain is stabilized for each spectrum using the gamma-ray peaks indicated in Table II.

Following the acquisition of each spectrum, the energy resolution and gain are evaluated from the data. A reference peak FWHM and centroid are determined and compared to limits specified by the program for these peaks. The reference peaks used in the resolution-gain checks are given in Table III along with optimum values for the energy resolution at count rates of 10 kHz.

The criterion for acceptable gain is that the reference peak centroid must fall within 1.5 channels of the channel specified for that peak. The acceptable resolution criterion is 15% greater than the optimized FWHM at 10 kHz for the reference peak.

TABLE I

ASSAY GAMMA RAYS

E_γ (keV)	Isotope
38.7	^{239}Pu
43.5	^{238}Pu
45.2	^{240}Pu
51.6	^{239}Pu
88.0	^{109}Cd
121.1	^{75}Se
122.1	^{57}Co
129.3	^{239}Pu
136.0	^{75}Se
148.6	^{241}Pu
152.7	^{238}Pu
279.5	^{75}Se

TABLE II

STABILIZATION GAMMA RAYS

	E_γ (keV)	Isotope
Zero	88.0	^{109}Cd
Gain	279.5	^{75}Se
	136.3	^{57}Co
	208.0	^{241}Pu[a]

[a] Parent of ^{237}U and ^{241}Am whose disintegrations give rise to this gamma ray.

TABLE III

GAMMA-RAY RESOLUTION AT 10 kHz

Isotope	E_γ (keV)	FWHM (keV)
^{109}Cd	88.0	0.49
^{57}Co	122.1	0.54
^{239}Pu	129.3	0.55
^{75}Se	279.5	0.75

2. Measurement Control. Measurement control data are the daily background, ^{75}Se, and ^{57}Co spectra acquired without a sample in the measurement well. Tests are applied to these data to verify proper performance of the instrument. (Other information, such as unattenuated intensities of the transmission gamma-ray peaks, is stored in a file that is accessed later during the measurements of plutonium solution samples.) In addition to the measurements made without a sample in the well, a complete densitometry analysis is performed on a plutonium reference foil. The reproducibility of the plutonium foil thickness is an assurance of the reproducibility of the plutonium concentration measurement capability for solution samples of well-defined thickness.

Acquisition times during measurement control are computer-controlled to provide 0.2% counting statistics in the peaks of interest. A source that is too weak to produce the required statistical precision within 1500 seconds is flagged for replacement. These precautions ensure a relatively constant contribution of the unattenuated intensities to the precision of the measured concentration.

The background spectrum during measurement control is obtained with the transmission sources positioned as for a passive assay (sources shielded from the detector). The background spectrum is acquired for two purposes: (1) to verify the absence of large, interfering backgrounds caused by the presence of external gamma-ray sources or, perhaps, contamination of the measurement well;

and (2) to measure the background net peak areas of all the assay gamma-ray peaks. These are stored in a data file and subtracted from the net peak areas obtained in all successive (measurement control and sample assay) measurements.

The ^{75}Se and ^{57}Co measurement control spectra are acquired to obtain the unattenuated intensities (net peak areas in the absence of sample attenuation) of the gamma-ray lines required for transmission measurements. These gamma-ray lines are the 121.1- and the 122.1-keV lines of ^{75}Se and ^{57}Co, respectively, used in the K-edge assay, and the 122.1-keV line of ^{57}Co along with the 136.0- and 279.5-keV lines of ^{75}Se, used to compute self-attenuation corrections in the isotopics assay.

In addition to the resolution and gain checks applied to the ^{75}Se and ^{57}Co measurement control spectra, a detector relative efficiency check is performed using the 121.1- and 136.0-keV net peak areas. The ratio of these two peak areas should remain constant for a given ^{75}Se source if the relative detection efficiency remains unchanged. The empirical calibration parameters for the isotopics assay are the relative detection efficiencies in this same energy region. (Refer to Sec. II.C.3.) Therefore, the relative efficiency check with these ^{75}Se gamma-ray lines is a measurement control procedure that applies to the isotopics assay. The check requires the measured ratio to be within 4σ of the stored value, where σ, the uncertainty in the measured ratio, is typically 0.2%.

At the completion of the measurement control sequence, the net peak areas obtained in the three (background, ^{75}Se, and ^{57}Co) spectra are stored in a data file along with the date and time of data acquisition. When accessed later for computation of transmissions, the unattenuated peak areas are corrected for decay of the ^{75}Se, ^{57}Co, and ^{109}Cd sources.

3. Sample Measurements. Assay of samples for plutonium concentration and isotopic fractions proceeds following the successful completion of the daily measurement control sequence. The working standard plutonium foil, mounted inside a standard solution sample cell, is inserted into the well and assayed by the K-edge technique using procedures identical to those used for assay of solution samples.

The solution sample assay consists of the ^{75}Se and ^{57}Co transmission measurements followed by the passive assay. The sample transmission and passive spectra are subjected to the checks on gain and resolution described previously. The count times are specified by the operator.

C. Analysis

1. Introduction. The reduction and analysis of spectral data to give plutonium concentration and isotopic fractions is automated and under computer control. Following checks on gain and resolution, net peaks areas are determined. All peaks areas used for the computation of transmissions are normalized to the ^{109}Cd 88-keV peak area. The next step in the data reduction is subtraction of net backgrounds from net peak areas to give net background-subtracted peak areas, A(E), where E corresponds to the gamma-ray energy.

Gamma-ray transmission at energy E, T_E, is defined as

$$T_E = \frac{A(E)}{A_0(E)} \quad ,$$

where $A_0(E)$ is the peak area in the absence of a sample, corrected for transmission source decay.

2. Concentration Assay. The K-edge assay uses the measured transmission ratio,

$$R = T_{122}/T_{121} \quad ,$$

to determine the plutonium concentration, ρ, using the equation:

$$\rho = -\ell n\, R/k \quad . \tag{1}$$

The origin of this relationship is discussed elsewhere[2-4,6,7,9] in detail. The constant, k, determined empirically using measured values of R obtained with solutions of known ρ, is equivalent to the product, $\Delta\mu x$, where $\Delta\mu$ is the change in the plutonium mass attenuation coefficient between the two energies (121.1- and 122.1-keV) and x is the sample thickness in the transmission path. The sample thickness for the Tokai cell is approximately 2 cm. (This is specified only approximately because the cell has a slight taper and the vertical position of the transmission path in the tapered cell is known only approximately.) The value of $\Delta\mu$ at the K-edge of plutonium is 3.4 cm^2/g. Therefore, the expected value of k for these measurements should be approximately 6.8 cm^3/g.

3. Isotopics Assay. The experimental results used to evaluate the plutonium isotopic ratios are the ratios of peak areas obtained from the passive gamma-ray spectrum. These ratios are converted to isotopic ratios, all relative to ^{239}Pu. The ^{239}Pu isotopic fraction is obtained from the inverse of the sum of the five isotopic ratios. Each isotopic ratio and the ^{239}Pu isotopic fraction are used to obtain the isotopic fractions of ^{238}Pu, ^{240}Pu, ^{241}Pu, and ^{242}Pu, respectively. The procedures for analysis are summarized below.

Two regions of the passive gamma-ray spectrum are used to obtain the peak area ratios. The high-energy region includes the 129-, 149-, and 153-keV gamma rays of ^{239}Pu, ^{241}Pu, and ^{238}Pu, and the low-energy region includes the 39-, 43-, 45-, and 52-keV gamma rays of ^{239}Pu, ^{238}Pu, ^{240}Pu, and ^{239}Pu, respectively. (Refer to Table I.) The computation of the isotopic ratios from the respective peak area ratios is different in the two energy regions. The isotopic ratios and isotopic fractions are all computed as weight fractions.

The weight fractions, f_I, for ^{238}Pu and ^{241}Pu are computed relative to f_{239} from the high-energy data using the expressions

$$\left(\frac{f_{238}}{f_{239}}\right)_H = \frac{A(153)}{A(129)} \cdot \frac{CF(153)}{CF(129)} \cdot R_{\lambda MB}\left(\frac{238}{239}\right) \cdot a = (R_{238})_H \qquad (2)$$

and

$$\frac{f_{241}}{f_{239}} = \frac{A(149)}{A(129)} \cdot \frac{CF(149)}{CF(129)} \cdot R_{\lambda MB}\left(\frac{241}{239}\right) \cdot b = R_{241} \quad . \tag{3}$$

The A and CF quantities are net peak areas (defined in Sec. II.C.1) and correction factors (for self-attenuation) for the peaks whose energies are indicated in parentheses. The correction factors are determined from the measured transmissions of the 122-, 136-, and 279-keV gamma-ray lines and by a numerical integration procedure described in Appendix A. The $R_{\lambda MB}$ ratios are determined by constants defining half-lives, atomic masses, and branching ratios for the two parent nuclei, as follows:

$$R_{\lambda MB}\left(\frac{238}{239}\right) = \frac{\lambda_{239}}{\lambda_{238}} \cdot \frac{M_{238}}{M_{239}} \cdot \frac{B_{129}}{B_{153}}$$

and

$$R_{\lambda MB}\left(\frac{241}{239}\right) = \frac{\lambda_{239}}{\lambda_{241}} \cdot \frac{M_{241}}{M_{239}} \cdot \frac{B_{129}}{B_{149}} \quad ,$$

where λ, M, and B are the decay constant, atomic mass, and branching ratio, respectively. These $R_{\lambda MB}$ values are given in Appendix B. The quantities a and b in Eqs. (2) and (3) are the relative detection efficiencies of the two passive gamma-ray lines. These quantities are determined empirically from measurements of reference solutions with well-defined isotopic content. The a and b values are stored in a data file that is accessed by the program.

$(R_{238})_H$ is one of two experimental values of the f_{238}/f_{239} ratio. The other, $(R_{238})_L$, is evaluated from the low-energy data as described below. The two are averaged, weighted by their respective uncertainties, to obtain R_{238}.

The ratios of the ^{238}Pu and ^{240}Pu isotopic fractions to the ^{239}Pu isotopic fractions, obtained from the low-energy data, can be expressed as

$$\left(\frac{f_{238}}{f_{239}}\right)_L = \frac{A(43)}{A(52)} \cdot \frac{R_{eff}(52)}{R_{eff}(43)} \cdot R_{\lambda MB}\left(\frac{43}{52}\right) \cdot K_{43} = (R_{238})_L \qquad (4)$$

and

$$\frac{f_{240}}{f_{239}} = \frac{A(45)}{A(52)} \cdot \frac{R_{eff}(52)}{R_{eff}(45)} \cdot R_{\lambda MB}\left(\frac{45}{52}\right) \cdot K_{45} = R_{240} \ . \qquad (5)$$

Here, the areas of the 43.5-, 45.2-, and 51.6- keV gamma-ray lines [A(43), A(45), and A(52), respectively] determine the relative amounts of ^{238}Pu, ^{240}Pu, and ^{239}Pu. The quantities $R_{eff}(43)$, $R_{eff}(45)$, and $R_{eff}(52)$ are the relative total efficiencies (including corrections for gamma-ray self-attenuation) for counting the corresponding gamma-ray lines. The $R_{\lambda MB}$ ratios are determined by constants that define the half-lives, branching ratios, and atomic masses for the two parent nuclei as follows:

$$R_{\lambda MB}\left(\frac{43}{52}\right) = \frac{\lambda_{239}}{\lambda_{238}} \cdot \frac{M_{238}}{M_{239}} \cdot \frac{B_{52}}{B_{43}}$$

and

$$R_{\lambda MB}\left(\frac{45}{52}\right) = \frac{\lambda_{239}}{\lambda_{240}} \cdot \frac{M_{240}}{M_{239}} \cdot \frac{B_{52}}{B_{45}} \quad ,$$

where λ, M, and B are literature values of the decay constants, atomic masses, and branching ratios, respectively. The constants, K_{43} and K_{45}, are empirical factors that correct the values of $R_{\lambda MB}$ determined using literature values. The $R_{\lambda MB}$ values are compiled as constants in the program. The values of K_{43} and K_{45} are stored in a data file that is accessed by the program. The $R_{\lambda MB}$ values are given in Appendix C.

The relative efficiencies, R_{eff}, are obtained for each measurement by an interpolation process using data from the passive spectrum. The relative efficiency for counting a gamma-ray line at energy E from a given isotope is defined as

$$R_{eff}(E) = A(E) \div B_E \quad ,$$

where A(E) and B_E are the net peak area and branching ratio of the gamma ray at energy E. The net peak areas for the two gamma-ray lines at 38.7 and 51.6 keV from ^{239}Pu are used to establish the energy dependence of R_{eff} in the energy region between these two gamma-ray lines. The interpolation is performed with the assumption that $\ell nR_{eff}(E)$ is a linear function of $\ell n(\ell nE)$ such that $\ell nR_{eff}(E)$ vs $\ell n(\ell nE)$ plotted for E values of 38.7 and 51.6 keV establish a slope and intercept over this energy region.

Measurements with solution standards of well-determined isotopic content are used to establish the constants, K_{43} and K_{45}. These constants are very near unity because they correct for small errors in the literature values for decay constants and branching ratios.

Because of the absence of a ^{242}Pu gamma-ray line of sufficient intensity for an experimental measurement of R_{242}, isotopic correlation techniques are used to determine R_{242}. The correlation used is

$$R_{242} = \frac{f_{242}}{f_{239}} = R_{240} \cdot (C_2 + C_1 \cdot R_{240}) \quad . \qquad (6)$$

The values of the constants C_2 and C_1 used to analyze the isotopics data initially were obtained* from a fit to pressurized-water reactor (PWR) and boiling-water reactor (BWR) data.

The five isotopic weight ratios, R_{238} [obtained from the weighted average of $(R_{238})_H$ and $(R_{238})_L$], $R_{239} \equiv 1$, R_{240}, R_{241}, and R_{242}, are combined to give the ^{239}Pu isotopic weight fraction:

$$f_{239} = (1 + R_{238} + R_{240} + R_{241} + R_{242})^{-1} \quad . \qquad (7)$$

---

*These results were obtained by S. T. Hsue, Los Alamos National Laboratory Group Q-1.

The weight fractions of the other isotopes, f_I, are determined by

$$f_I = R_I \cdot f_{239} \quad . \tag{8}$$

III. EVALUATION PROCEDURES

A. Samples

A total of 133 process solution samples were analyzed by the Tokai instrument between October 1979 and February 1981. The results were compared to the destructive analytical results obtained at Tokai with the same samples.

Eight- to ten-milliliter volumes of solution were withdrawn from the process into plastic vials used for transport of solution samples throughout the Tokai facility. The solution samples originated either from the freshly separated product vessel or from the aged product storage vessel. Originally, certain of these samples were set aside for nondestructive analysis, whereas others, withdrawn from the process at approximately the same time, were used for destructive assay. Because the destructive assay results were the reference values for the nondestructive assay results, it was desirable to minimize the sampling errors. Therefore, the sampling procedures were modified after February 1980 so that six discrete samples withdrawn from the process were initially combined before distribution. One-half of the total volume was separated and used for destructive analysis. The other half was distributed among the nondestructive assay stations.

The Tokai instrument requires a minimum solution volume of 7 ml for the K-edge assay. Because the collimation is enlarged for the passive count, approximately twice this volume is required for the isotopics assay. The volumes of the solution samples assayed by the Tokai instrument were, typically, 15 ml. The plutonium concentrations of these samples were, typically, between 200 and 250 g/ℓ.

The americium content of the aged solution samples was as high as 0.2% in some cases. Because of the excessively high detector count rates ($>10^5$ Hz) from ^{241}Am in the passive assay configuration, the aged samples cannot be adequately analyzed for isotopic content with the present Tokai instrument. Therefore, the isotopic fractions are reported only for those samples (64 in number) from which the americium was freshly separated.

B. Reference Values

1. Concentration. The destructive analytical procedure for determination of plutonium concentration is a ceric back-titration to a potentiometric end point. The density of the plutonium product solution is determined precisely. This solution is sampled (for 10 to 25 mg of plutonium) and diluted with 2 ml of 5$\underline{N}$ nitric acid. The valence is adjusted to Pu(VI) by mixing for 5 to 10 minutes with a strong oxidizing agent (AgO). Nitrates are removed from the mixture by the addition of 5 ml of 6$\underline{N}$ sulfuric acid followed by 0.5 g of powdered sulfamic acid. A known volume (5 ml) of Fe(II) standard solution (as $FeSO_4$) is introduced in excess by calibrated pipette to reduce the plutonium to Pu(IV). The Fe(III) is back-titrated to Fe(II) with a standard Ce(IV) solution to the potentiometric end point. The Fe(II) solution (∿0.05$\underline{N}$) is standardized by potentiometric titration of pipetted 5-ml samples with a standard Ce(IV) solution. Standardization of the (∿0.025$\underline{N}$) Ce(IV) solution is accomplished by addition of 3 ml of primary standard dichromate to pipetted 5 ml of Fe(II) solution and by back-titration of the resulting Fe(III) (standard) with the Ce(IV) solution.

Each unknown plutonium sample assay is preceded by standardization of the Fe(II) and Ce(IV) solutions. Each plutonium sample assay is performed in triplicate. A typical delay of 8 to 12 hours in the availability of these destructive analytical results is due primarily to the time required for performing measurement control and standardization procedures in advance of the plutonium sample titrations. An uncertainty of 0.5% (1σ), determined from the precision obtained from the triplicate measurements, is ascribed to the reference values used for comparison with the nondestructive assay results of plutonium concentration in the product solution samples.

2. Isotopics. Thermal ionization mass spectrometry is used to determine the mass fractions of the plutonium isotopes in the freshly separated process solution samples. The mass spectrometer uses a single-focusing 90° sector with a 305 mm radius.

Anion exchange methods for separation of uranium and americium from the plutonium are used routinely in preparation of the freshly separated samples for mass spectrometry. The americium content of these samples is too low to be of concern. However, uranium (primarily as ^{238}U) is present at levels of approximately 0.25 parts in 10^3 (U:Pu) in the plutonium product solution. The evaporation temperature is significantly lower for plutonium than uranium so that the effects of contamination (of ^{238}Pu by ^{238}U) can be reduced by careful control of the filament current. However, the ^{238}U is a low-level environmental contaminant (of the filament, of solution containers, etc.) that appears as an interference even if ion exchange procedures are used routinely. Thus, the mass spectrometry techniques will not provide the precision required of the ^{238}Pu reference value (<0.5%) for evaluation of the nondestructive assay technique. The typical precisions of the mass spectrometry technique (applied to high-burnup samples) for assay of the other plutonium isotopes (0.05% for ^{239}Pu, 0.2% for ^{240}Pu, 0.4% for ^{241}Pu, and <1% for ^{242}Pu) are the estimated uncertainties in the respective reference values used for comparison with the passive assay results.

C. Nondestructive Measurements

The typical delay between the availability of a solution for nondestructive assay with the Tokai instrument and execution of the assay is approximately one-half hour. This time is used for various glovebox bagout procedures as well as for transfer of solutions into plastic measurement vials. Live count times for the ^{75}Se and ^{57}Co measurements were 1000 seconds (each), and 2000-second count periods were used for the passive assay. This corresponds to clock times of approximately 40 minutes each for the K-edge and passive assays.

Each sample was initially assayed in a single cycle. At some later time (usually within 24 hours of the original assay) the sample was repositioned in the well for a repeated, 10-cycle assay. This procedure permitted evaluation of the random contribution to the measurement precision and also provided the opportunity to evaluate the effects of sample repositioning. The data obtained in October 1979 as well as the data obtained in April 1980 were exceptions in that the assay protocol described above (single-cycle assay followed by a 10-cycle assay) was not observed. For the October 1979 assays the solution samples were provided by the analytical laboratory for the purposes of calibration of the instrument. For the April 1980 assays, the measurements were performed as a demonstration procedure. All other samples were assayed in the course of a process campaign or during other facility-related operations.

IV. DATA AND MEASUREMENT RESULTS

A. Transmission Spectra

The ^{75}Se and ^{57}Co transmission spectra for representative freshly separated (hereafter referred to as "fresh") and aged product solution samples are shown in Figs. 6 and 7, respectively. Only the transmission assay gamma rays are labeled. The portions of the 4096-channel spectra displayed in these figures correspond to the 35- to 160-keV energy range (approximately one-half of the full spectrum). This range includes all of the plutonium passive assay gamma rays as well as the transmission gamma rays used for the K-edge assay. Note that the intensities of the passive assay gamma rays relative to the intensity of the ^{109}Cd 88.0-keV gamma ray are quite small; this is due to restrictive collimation of the sample during the transmission measurements. Because of this collimation, it is possible to perform transmission measurements on the aged samples despite the presence of high levels of americium. Most of the structure that is prominent in the "aged" spectra and not in the "fresh" spectra is due to ^{241}Am activity.

Certain gamma-ray peaks appear in the fresh and not in the aged spectra. The most prominent example in Figs. 6 and 7 is the peak at 106.1 keV. This peak and numerous others are due to gamma rays from the decay of ^{239}Np. This short-lived ($t_{1/2}$ = 2.35 d) isotope is present because neptunium accompanies plutonium in the process. The ^{239}Np is the daughter of the ^{243}Am; americium is separated from plutonium during the reprocessing. Therefore, the ^{239}Np

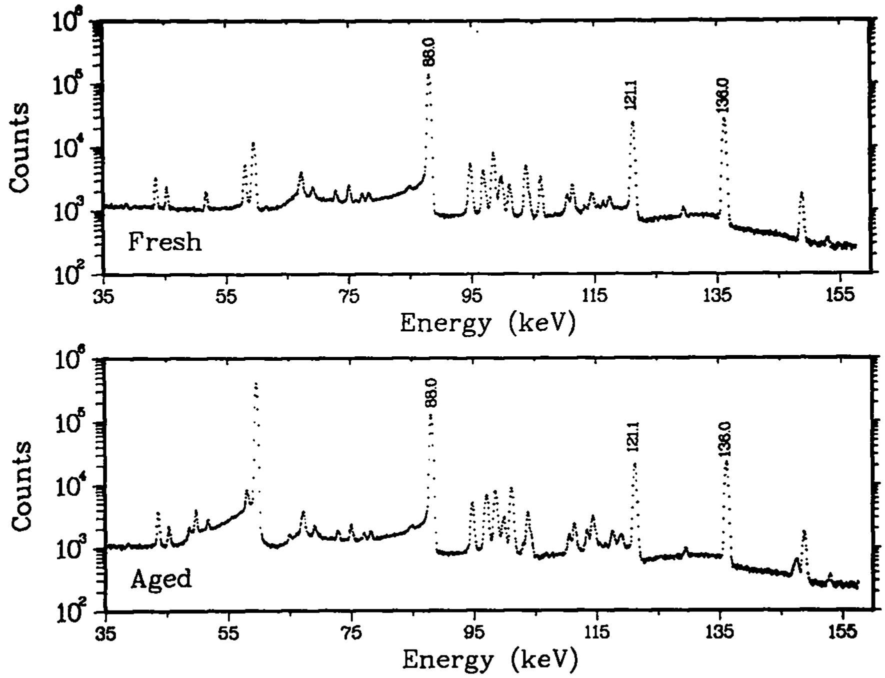

Fig. 6.
The ^{75}Se transmission spectra, obtained in 1000-second counting periods with fresh and aged product solution samples, in the energy region between 35 and 160 keV. This energy range includes all of the assay gamma rays except the ^{75}Se gamma ray at 279.5 keV. The energies (in keV) of the ^{109}Cd and ^{75}Se gamma rays used for assay are noted above the peaks.

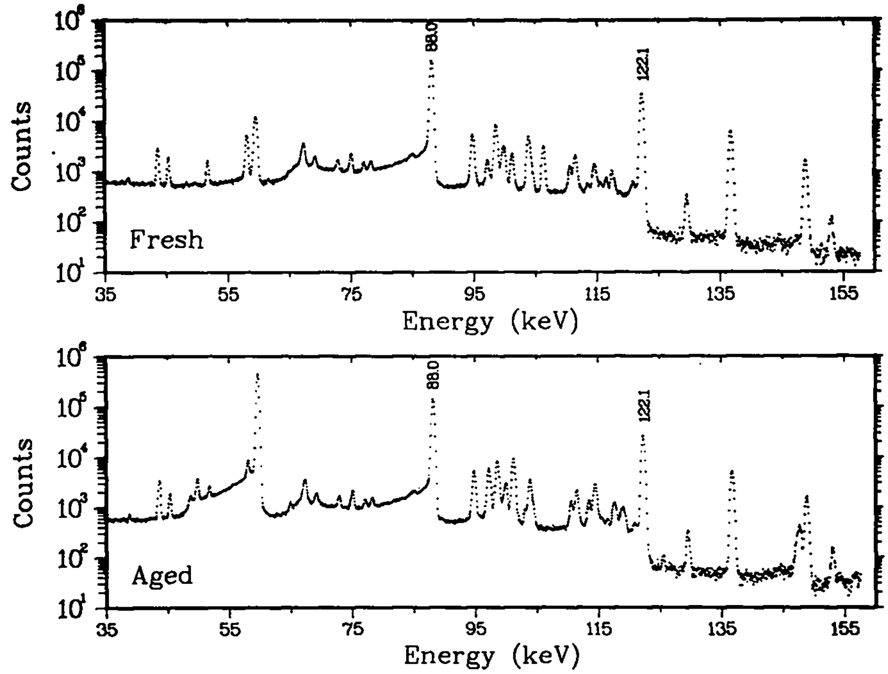

Fig. 7.
The ^{57}Co transmission spectra, obtained in 1000-second counting periods with fresh and aged product solution samples, in the energy region between 35 and 160 keV. This energy range includes all of the assay gamma rays except the ^{75}Se gamma ray at 279.5 keV. The energies (in keV) of the ^{109}Cd and ^{57}Co gamma rays used for assay are noted above the peaks.

activity is negligible in the product samples 2 weeks after processing. Only one of the ^{239}Np gamma rays has any significance (as a possible interference) to the nondestructive assay; the energy of this gamma ray is 277.6 keV. The peak is prominent in the upper sixth of the ^{75}Se transmission spectra for fresh samples. Figure 8 shows this region for both fresh and aged samples. The ^{75}Se gamma ray at 279.5 keV is the only assay gamma ray that is outside the 35- to 160-keV region displayed in Figs. 6 and 7. It is one of three gamma rays (the other two being the ^{75}Se 136.0-keV gamma ray and the ^{57}Co 122.1-keV gamma ray) whose transmissions are used to compute the correction factors for the passive gamma-ray self-attenuation in the 130- to 150-keV region. The maximum contribution of the 277.6-keV peak to the 279.5-keV net peak area is less than 1%. The effect on the correction factors in the 130- to 150-keV region resulting from a 1% change in the 279.5-keV transmission is approximately 0.02%; it is, therefore, not significant.

B. Measurement Control (Plutonium Foil) Results

Figure 9 is a summary of the measurement control results obtained from the K-edge assay of the plutonium foil. The effective concentration (in g/ℓ) is plotted vs date for the time period beginning with the date the foil was available for measurement at PNC (approximately 2 months after solution sample measurements began). The open points in the figure correspond to a single (40-minute) K-edge assay. The solid points are the average results for 10 or more such assays performed in a continuous sequence. The standard deviation in the solid points is 0.23% for the 1-year period. The statistical contribution to these data is 0.18%. The residual component of 0.14% is in reasonable agreement with a previous evaluation of the random (other than statistical) contribution (0.13%) determined[9] from repeated measurements with very long count times.

The data indicate a slight tendency of the foil assay result to decrease with time. A linear fit to the solid points in Fig. 9 shows a decrease of 0.2% per year, although the standard deviation in these data relative to the fit (0.22%, 1σ) is only a small improvement over that relative to the average thickness (0.23%). Hence a very large uncertainty is ascribed to this measured decrease.

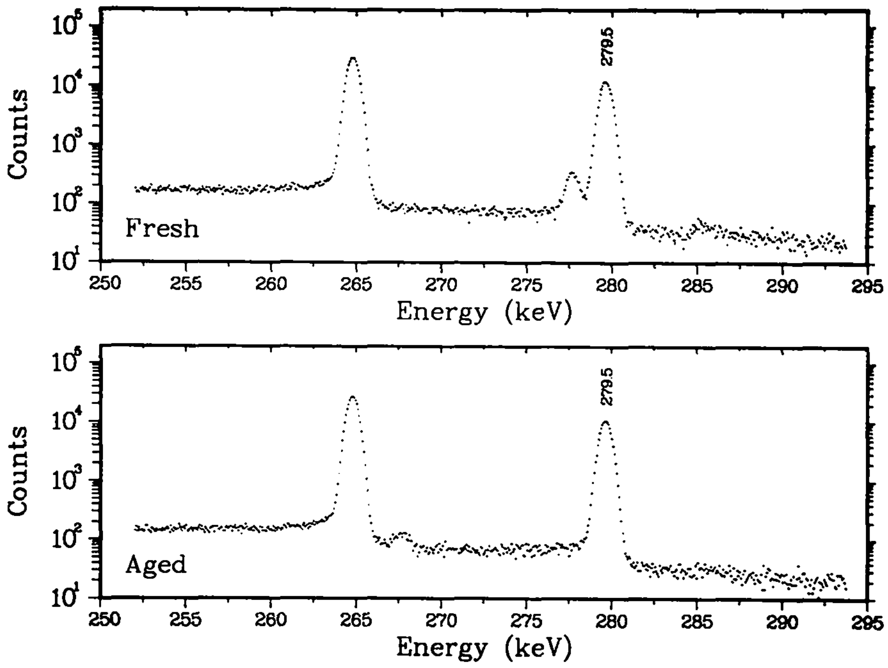

Fig. 8.
The upper sixth of the ^{75}Se spectra, obtained in 1000-second counting periods with fresh and aged product solution samples, which includes the ^{75}Se gamma ray at 279.5 keV.

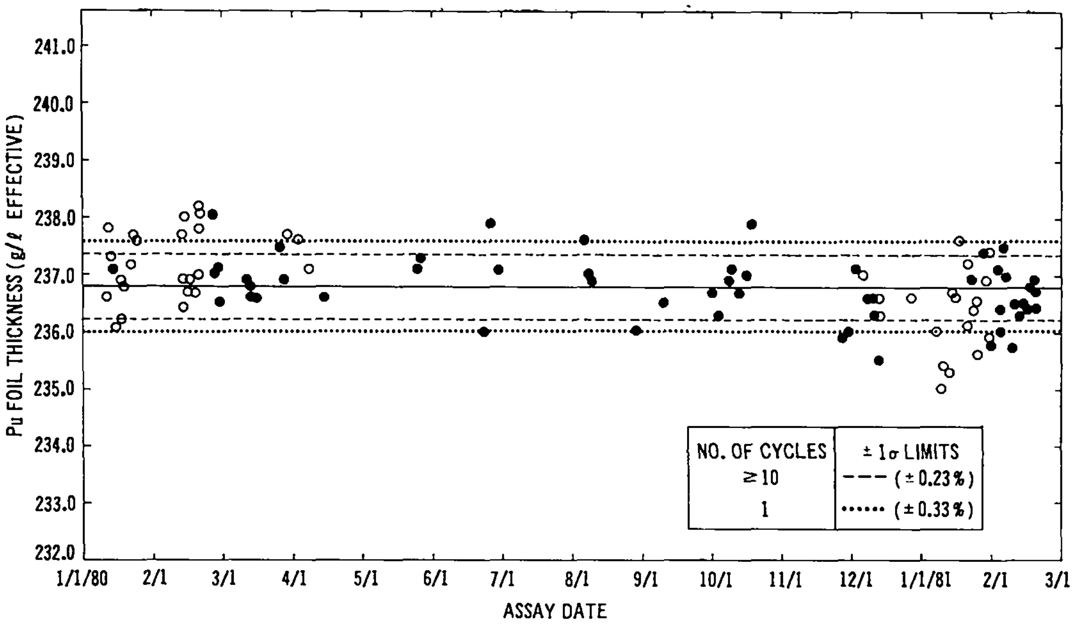

Fig. 9.
The K-edge assay results (in equivalent g Pu/ℓ) for the plutonium foil, plotted vs date. The open circles correspond to a single (40-minute) assay. The solid points are the average results of 10 to 15 repeated assays. The solid line is the mean result of 236.8 g/ℓ, which is the same for single and multiple assays. The dashed and dotted lines indicate the standard deviations (1σ) for the multiple- and single-assay results, respectively.

Several effects may contribute to a change with time in the measured foil thickness; all are related to the decay of ^{241}Pu to ^{241}Am; all contribute to a decrease in the measured foil thickness. The most obvious effect is the loss of plutonium thickness (0.05% per year), resulting from the decay of ^{241}Pu. Another effect is the increase in intensity of the weak ^{241}Am peak at 123.0 keV; this increases the apparent transmission at 122.1 keV and reduces the assay result by an amount estimated to be less than 0.1% per year. A third effect is the growth of the pileup peak at 119.1 keV (resulting from the increase in intensity of the 59.5-keV gamma ray of ^{241}Am). Although the pileup peak is small and considerably removed (two times the FWHM) from the low-energy background region of the ^{75}Se 121.1-keV peak, it is possible that a very small reduction in the effective transmission at 121.1 keV and, hence, a very small (less than 0.05%) reduction in the assay can result from the pileup. Other possible factors (such as the reduced background caused by the 125.0-keV K-edge of americium) have been ruled out.

The radioactivity of the working reference foil appears to affect the K-edge assay result for this foil. However the effects (both observed and predicted) are small enough in the 1-year period so that the foil can be used to validate the reproducibility of the plutonium concentration measurement capability for solutions of a given thickness. For a longer period, a nonradioactive working reference might prove to be more effective for the purposes of measurement control.

C. Plutonium Concentration Results

The K-edge assay results for plutonium solution samples are discussed relative to the (reference) results of destructive analysis performed on these samples. The assays are summarized in Fig. 10, which shows the relative results (the percentage deviation of the K-edge assay from the reference value) plotted vs sample number. The results are shown for 10 sets of samples, starting with those made available in October 1979 for the purposes of calibration. The concentration results (K-edge and reference values) for each of the 133 samples are tabulated in Appendix D.

The plotted data points are obtained from the average of the single K-edge assay result and the 10-cycle average for each sample. The standard deviation in the relative data in each sample set along with the average result for each sample set are shown as dashed and solid lines, respectively, and are also

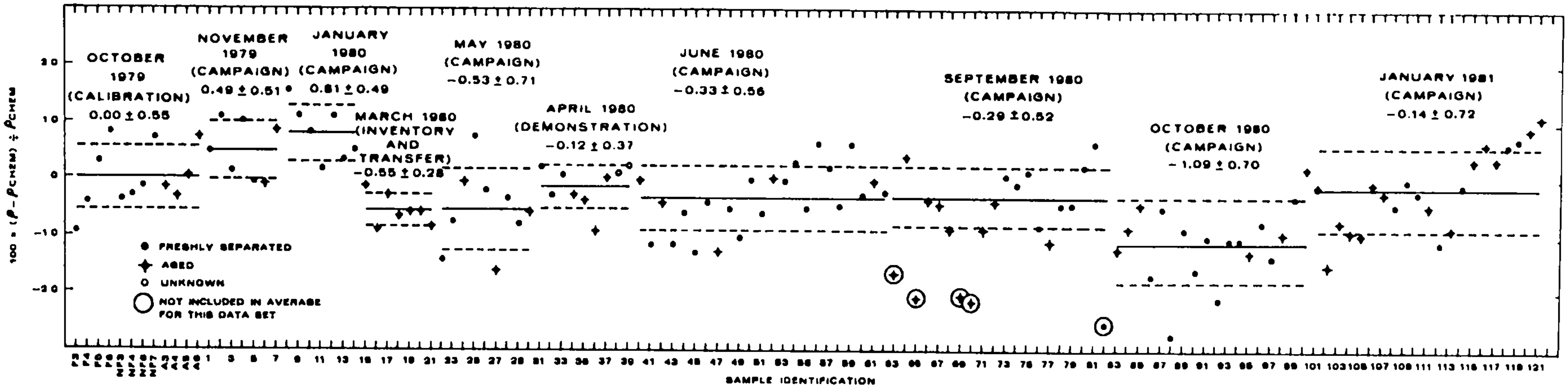

Fig. 10

The relative K-edge assay results for the solution samples, plotted vs sample number. (The relative result is the percentage deviation from the destructive analytical result obtained for the same sample.) The average relative result is shown for each sample set as a solid line. The standard deviation (1σ) in the data for each sample set is shown by the dashed lines. The average results and standard deviations are indicated above each data set. Different symbols are used for fresh and aged samples. Five data points excluded from the calculations of the average and the standard deviation (refer to text) are circled.

indicated above the individual data sets in Fig. 10. The fresh and aged samples are distinguished with different symbols. Five data points from the September 1980 sample set are circled in Fig. 10; these data points deviate by more than two standard deviations from the mean. Furthermore, nearly simultaneous assays of solutions obtained from the same set of samples using a different nondestructive technique based on high-resolution (absolute) counting of passive gamma rays of plutonium (Task H of the TASTEX Program[1]) showed similar discrepancies for these five samples relative to the mean result. Therefore the results for these five samples (numbers 63, 65, 69, 70, and 82) have been eliminated from the data evaluation to avoid a bias that appears to be independent of the K-edge measurements.

The results prior to February 1980 and those of the October 1980 campaign show discrepancies from the trend observed in Fig. 10 for the remaining data. The change instituted in the sampling procedures after February 1980 may account for these earlier discrepancies. However, there is no such cause to explain the low average for the relative results of October 1980. Furthermore, the spectra for the October 1980 samples appear free from unusual structure or background, and the measurement control results (Fig. 9) show no evidence of discrepancies in the foil assay during this period. The K-edge evaluation has been carried out using the complete data set [row (i), Table IV] as well as the data set excluding measurements prior to February 1980 [row (ii)] and the data set since February 1980 and excluding the October 1980 results [row (iii)].

One of the measurement uncertainties examined during the evaluation at Tokai is the contribution to the measurement precision caused by repositioning of the sample cell. The cell has a taper that is large enough to cause a 0.25% change in the calibration parameter, $\Delta\mu x$, for a 1-mm change in vertical position. The repositioning of the cell between the single assay and the 10-cycle assay of each sample gave an average relative reproducibility ($\bar{\delta}$) for each sample set (Table IV). The contribution of the counting statistics to $\bar{\delta}$ varies between 0.2 and 0.3% (according to sample composition and source strength and according to whether the same set of unattenuated intensities was used to compute the single-cycle and the 10-cycle assay results). Row (i) of Table IV gives the average value of $\bar{\delta}$ for all the data (0.23%). Agreement between this result and the statistical estimate indicates that all other random effects, such as those that are due to routine sample positioning, are small and can be ignored for the purposes of this evaluation.

TABLE IV

PLUTONIUM CONCENTRATION RESULTS

Sample Set (number of samples[a])	Average Reproducibility (%)	Average Deviation From Reference Value (%)					
		ALL SAMPLES		FRESH SAMPLES		AGED SAMPLES	
	$\bar{\delta}\pm1\sigma_{\bar{\delta}}$	$\bar{\Delta}\pm1\sigma_{\bar{\Delta}}$	$(1\sigma_{data})$	$\bar{\Delta}\pm1\sigma_{\bar{\Delta}}$	$(1\sigma_{data})$	$\bar{\Delta}\pm1\sigma_{\bar{\Delta}}$	$(1\sigma_{data})$
Oct. '79 (8f,4a)	n.a.	0.00±0.15	(0.55)	-0.04±0.21	(0.59)	0.01±0.21	(0.43)
Nov. '79 (5f,2a)	0.22±0.07	0.49±0.19	(0.51)	0.52±0.24	(0.53)	0.40±0.50	(0.71)
Jan.'80 (7f,0a)	0.11±0.04	0.81±0.19	(0.49)	0.81±0.19	(0.49)	n.a.	
Mar.'80 (0f,7a)	0.26±0.06	-0.55±0.11	(0.28)	n.a.		-0.55±0.11	(0.28)
May'80 (6f,3a)	0.40±0.08	-0.53±0.24	(0.71)	-0.43±0.29	(0.70)	-0.70±0.47	(0.82)
Apr.'80 (3f,4a,2u)	n.a.	-0.12±0.13	(0.37)	0.00±0.15	(0.26)	-0.40±0.19	(0.37)
June'80 (18f,5a)	0.29±0.06	-0.33±0.12	(0.56)	-0.34±0.15	(0.61)	-0.32±0.21	(0.46)
Sept.'80 (8f,7a)	0.26±0.06	-0.29±0.13	(0.52)	-0.09±0.16	(0.44)	-0.53±0.19	(0.50)
Oct.'80 (12f,6a)	0.18±0.03	-1.09±0.17	(0.70)	-1.25±0.21	(0.74)	-0.75±0.23	(0.56)
Jan.'81 (7f,14a)	0.12±0.02	-0.14±0.16	(0.72)	-0.07±0.25	(0.65)	-0.19±0.21	(0.77)
(i) All Data (74f,52a,2u)	0.23±0.02[b]	-0.28±0.06	(0.73)	-0.23±0.09	(0.81)	-0.37±0.09	(0.61)
(ii) Since Jan.'80 (54f,46a,2u)		-0.44±0.07	(0.67)	-0.46±0.10	(0.74)	-0.44±0.09	(0.59)
(iii) Since Jan.'80 Excl. Oct.'80 (42f,40a,2u)		-0.29±0.06	(0.57)	-0.23±0.09	(0.58)	-0.39±0.09	(0.59)
(iv) $\overline{1\sigma_{data}}$ (weighted av.)							
(74f, 52a, 2u)			(0.58)		(0.59)		(0.56)
(42f, 40a, 2u)			(0.56)		(0.57)		(0.57)

[a] f = fresh; a = aged; u = unknown.
[b] (63f, 44a).

The average deviation from the reference value ($\overline{\Delta}$) is also given for each sample set in Table IV. Column 3 lists $\overline{\Delta}$ determined using all (fresh and aged) samples in the sample set (Fig. 10); columns 4 and 5 give the $\overline{\Delta}$ values for the fresh and aged samples separately. The values in parentheses ($1\sigma_{data}$) are the standard deviations in the data points for the sample set. (The $1\sigma_{data}$ values are also given in Fig. 10 for each sample set.) The uncertainties ($1\sigma_{\overline{\Delta}}$) are the standard deviations in the $\overline{\Delta}$ values (equivalent to $1\sigma_{data}$ divided by $\sqrt{N}$ where N is the number of samples). The values of $\overline{\Delta}$, $1\sigma_{\overline{\Delta}}$, and $1\sigma_{data}$ are also given at the bottom of Table IV for all data (128 samples), the data since January 1980 (102 samples), and the data since January 1980 excluding the October 1980 results (84 samples). Row (iv) gives the weighted average over the 10 sample sets of $1\sigma_{data}$ (that is, $\overline{1\sigma_{data}}$) where weighting is done according to the number of samples in the sample set.

Comparison of $\overline{\Delta}$ values for different data sets shows agreement (consistent with the uncertainties, $1\sigma_{\overline{\Delta}}$) among these quantities, with the exception of the results obtained prior to March 1980 and those for October 1980. When $\overline{\Delta}$ is evaluated for all samples [refer to rows (i) and (ii)], the standard deviation in the data ($1\sigma_{data}$) is larger than the average of this result over the individual data sets [$\overline{1\sigma_{data}}$, row (iv)], unless the results for October 1980 as well as those prior to March 1980 are omitted from the evaluation. [Refer to row (iii).]

For the data obtained prior to March 1980, some of the systematic effects that cause shifts in $\overline{\Delta}$ between sample sets might be a result of the sampling procedures used through February 1980. (Refer to Sec. III.A.) However, there is no obvious cause to cite for the discrepancy in the average deviation ($\overline{\Delta}$) for October 1980.

The possibility of small systematic effects in the K-edge assay of the foil, resulting from ^{241}Pu decay and ^{241}Am buildup (Sec. IV.B), motivates concern over the possibility of systematic differences in the K-edge assays of fresh and aged solutions. If such effects are significant, they should appear as differences in $\overline{\Delta}$ for the fresh and aged samples such that $\overline{\Delta}$ should be more negative for the aged samples. Columns 4 and 5 of Table IV tabulate the results for fresh and aged samples separately. Considering the results for the 10 sample sets, only those for April 1980, September 1980, and October 1980 show differences outside the $1\sigma_{\bar{\Delta}}$ limits. The significance of the difference is not large for the April 1980 data because the number of samples is small (application of the Fisher t-test ranks the difference at the 75% confidence level). The October 1980 discrepancy is opposite the expected trend (that is, $\bar{\Delta}$ is more negative for the fresh samples). Furthermore, the significance of this difference becomes greatly reduced (to the 70% confidence level) if the anomalously low result for fresh sample No. 88 (refer to Fig. 10) is omitted. The significance of the difference in $\bar{\Delta}$ for the aged and fresh samples of September 1980 (ranked at the 95% confidence level) could be the result of unusually high levels of americium in the aged samples. The americium content was higher by a factor of 2 to 3 than that of other aged samples assayed by the K-edge instrument.

The $\bar{\Delta}$ results for all samples differ for the fresh and aged samples at the 70% confidence level. This difference is well below the 10% confidence level for the samples assayed since the January 1980 campaign because of the substantial effect of the October 1980 data where the results for fresh and aged samples show the reverse trend. If the October 1980 data are omitted from the evaluation, the significance of the difference in $\bar{\Delta}$ for fresh and aged samples is, again, at the 70% confidence level. Seventy per cent is not sufficiently large to conclude that a systematic difference exists between the K-edge results for fresh and aged samples, on the average. However, the 95% result obtained for the September 1980 data indicates that for samples with very high americium content, the magnitude of the effect might be as large as 0.4%. On the average, the magnitude of the effect, if it does exist, is less than 0.2%, as indicated by the average values of $\bar{\Delta}$ in Table IV.

An average relative bias ($\bar{\Delta} \pm 1\sigma_{\bar{\Delta}}$) of -0.3% ± 0.06 is observed. This implies a need for a calibration adjustment if the reference concentrations are assumed to be accurate.

The contribution of the K-edge counting statistics to the standard deviation of the data points shown in Fig. 10 is 0.25%. This statistical contribution, which has been shown to represent the random contribution to the uncertainty in the K-edge assay, can be unfolded from the standard deviation in the relative results ($1\sigma_{data}$) to obtain an estimate of the combined accuracies of the destructive and nondestructive techniques, independent of

K-edge counting statistics. The unfolded result for all the data [row (i)] is 0.69%. Because the data prior to February 1980 were subject to sampling effects that no longer apply, it is appropriate to consider the standard deviation in the results obtained since January 1980 as more representative of the accuracies of the two analytical techniques. Unfolding the random contribution from this result [row (ii)] gives 0.62%. Furthermore, if the anomalous results of October 1980 are excluded [row (iii)], the limiting value of 0.51% is obtained for the combined accuracy of the two techniques. A contribution to this quantity of 0.1% (in quadrature) is ascribed to the observed systematic differences in the relative bias for fresh and aged samples.

D. Passive Spectra

The passive gamma-ray spectra from 35 to 160 keV are shown for representative BWR and PWR product solution samples in Fig. 11. The 88-keV ^{109}Cd gamma-ray line is labeled along with the passive plutonium gamma-ray lines used for the isotopics analysis (refer to Table I and Sec. II.C). No significant differences are observed in the passive spectra for BWR and PWR samples.

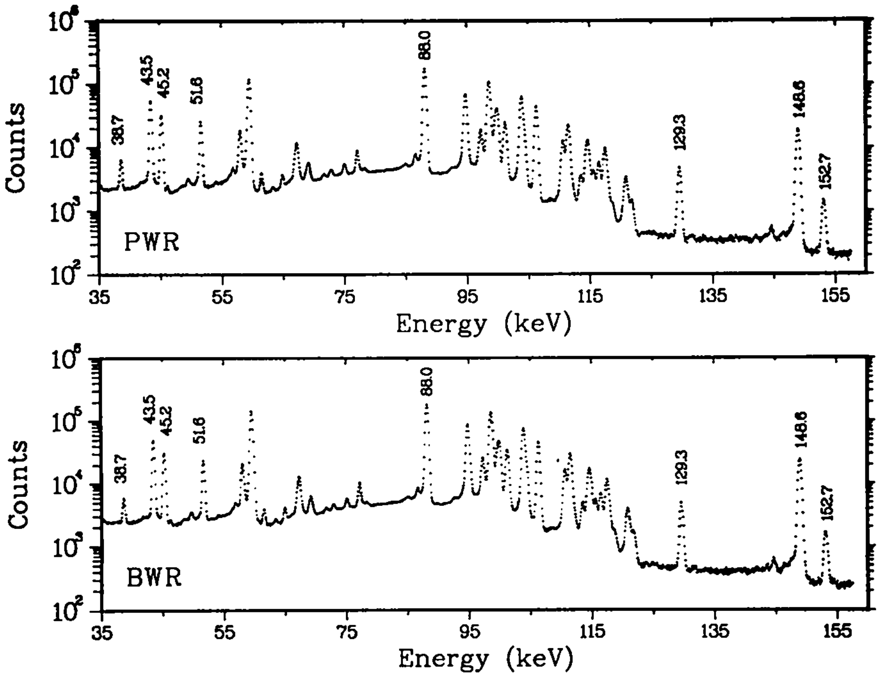

Fig. 11.
Passive gamma-ray spectra for BWR and PWR product solution samples, obtained in 2000-second counting periods, in the energy region between 35 keV and 160 keV. This energy region includes all of the passive assay gamma-ray lines. These are labeled, along with the 88-keV ^{109}Cd gamma-ray line.

E. Plutonium Isotopics Results

1. Introduction. Figures 12 through 16 show the results of the isotopics analysis of the passive data, plotted as the percentage deviation from the mass spectrometry result vs the ^{240}Pu isotopic fraction, for ^{242}Pu, ^{241}Pu, ^{240}Pu, ^{239}Pu, and ^{238}Pu, respectively. The results are shown for the fresh samples: 34 BWR (open symbols) and 30 PWR. Different symbols are used for each of the sample sets, which are distinguished by date. Each data point in Figs. 12 through 16 corresponds to the average isotopic fraction determined from 10 repeated assays (each consisting of a 2000-second transmission measurement and a 2000-second passive count) of the sample. Thus, the contribution of counting statistics to the uncertainty in the average result is small. The random contribution to the uncertainty in each sample assay has been evaluated for each isotope from the standard deviations in the 10-cycle data averaged for the 64 samples. These results, given in Table V, are comparable to the calculated contribution of counting statistics to the standard deviation. The statistical contribution to the uncertainties in the 10-cycle averages should be less than the values in Table V by a factor of $\sqrt{10}$.

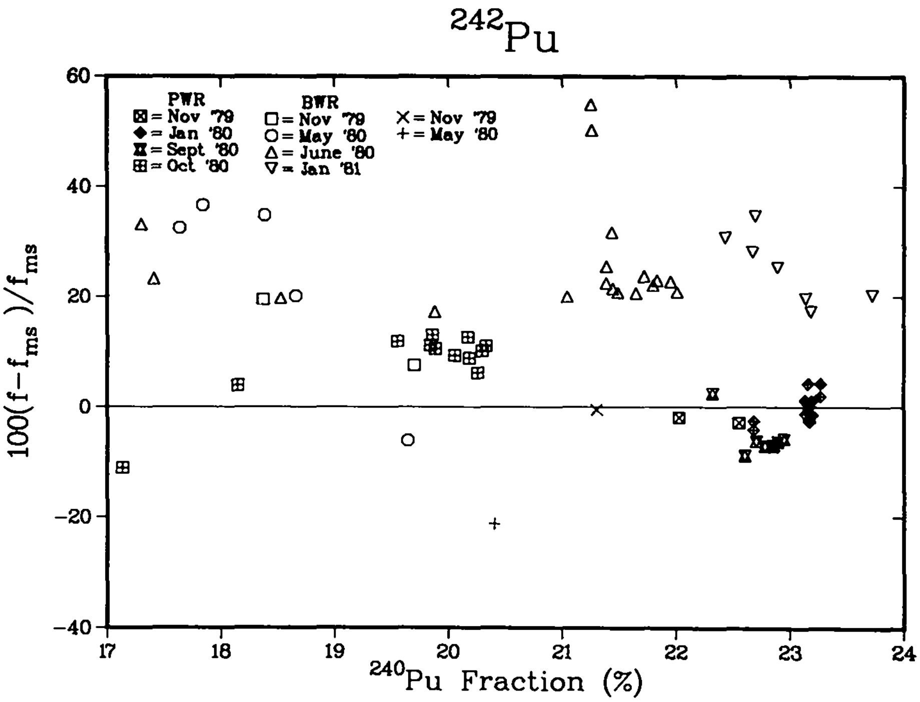

Fig. 12. Percentage difference between ^{242}Pu isotopic fraction determined by passive gamma-ray assay and mass spectrometry, plotted vs ^{240}Pu isotopic fraction for the fresh product solution samples. The analysis was performed with the original empirical parameters (Table VI). The open symbols distinguish the BWR from the PWR samples. Different symbols are used for different data sets.

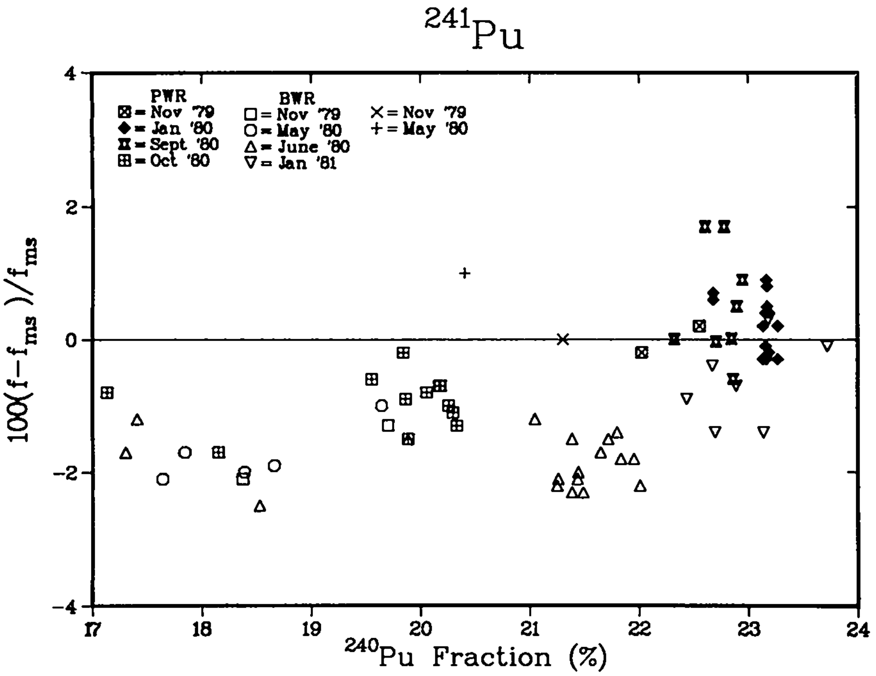

Fig. 13.
Percentage difference between ^{241}Pu isotopic fraction determined by passive gamma-ray assay and mass spectrometry, plotted vs ^{240}Pu isotopic fraction for the fresh product solution samples. The analysis was performed with the original empirical parameters (Table VI). The open symbols distinguish the BWR from the PWR samples. Different symbols are used for different data sets.

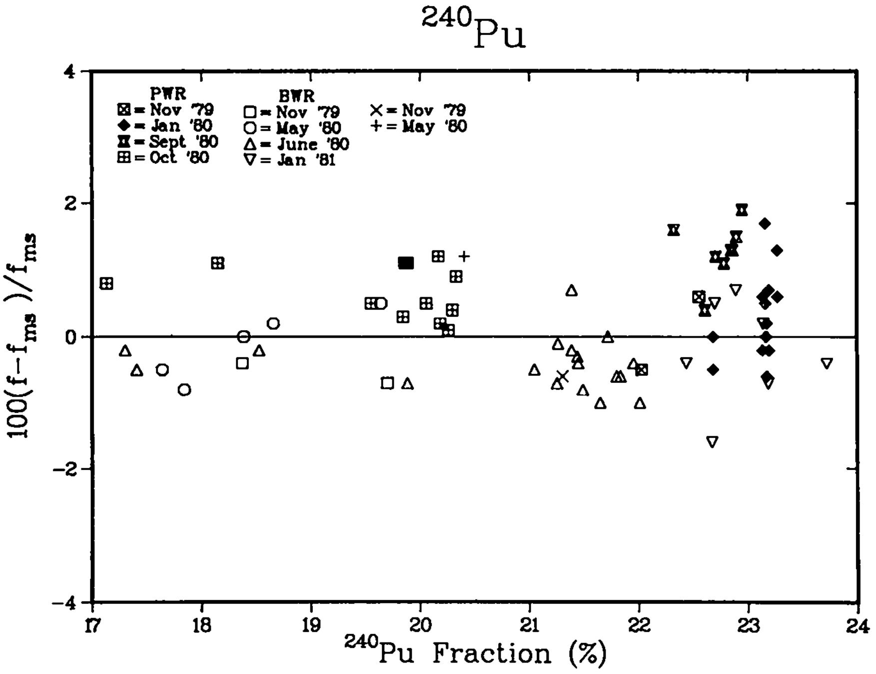

Fig. 14.
Percentage difference between ^{240}Pu isotopic fraction determined by passive gamma-ray assay and mass spectrometry, plotted vs ^{240}Pu isotopic fraction for the fresh product solution samples. The analysis was performed with the original empirical parameters (Table VI). The open symbols distinguish the BWR from the PWR samples. Different symbols are used for different data sets.

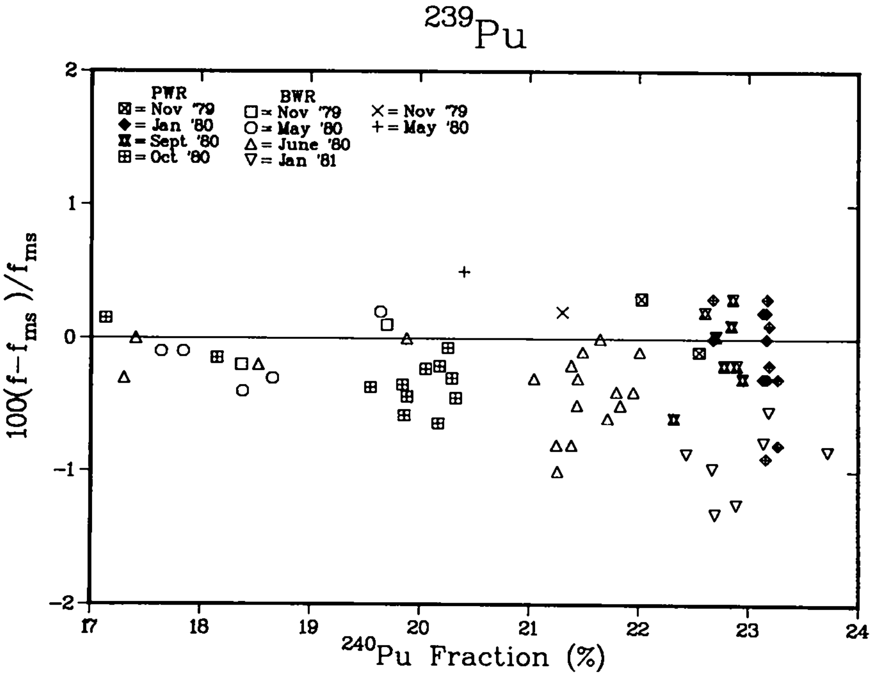

Fig. 15.
Percentage difference between ^{239}Pu isotopic fraction determined by passive gamma-ray assay and mass spectrometry, plotted vs ^{240}Pu isotopic fraction for the fresh product solution samples. The analysis was performed with the original empirical parameters (Table VI). The open symbols distinguish the BWR from the PWR samples. Different symbols are used for different data sets.

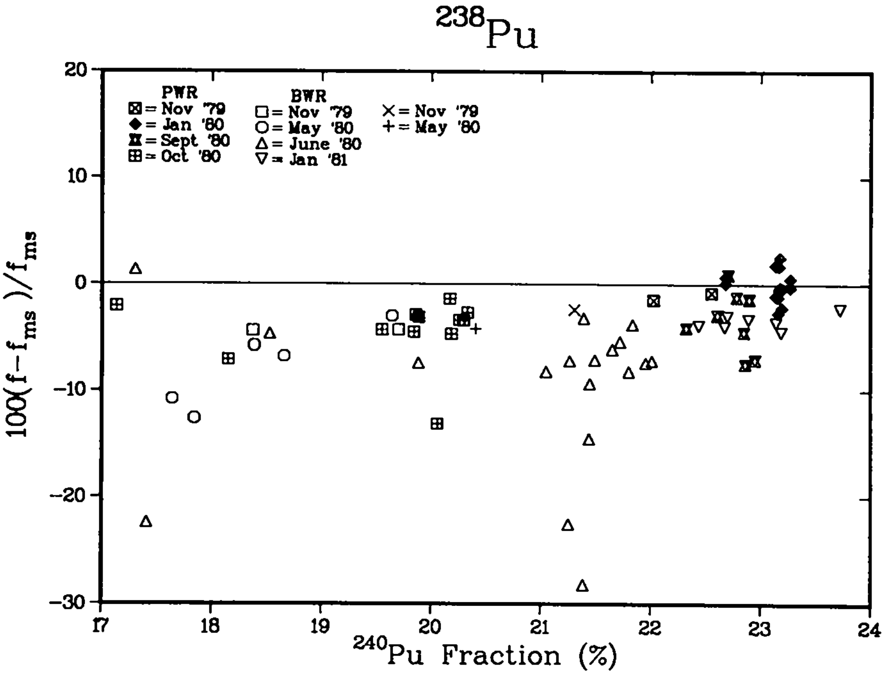

Fig. 16.
Percentage difference between ^{238}Pu isotopic fraction determined by passive gamma-ray assay and mass spectrometry, plotted vs ^{240}Pu isotopic fraction for the fresh product solution samples. The analysis was performed with the original empirical parameters (Table VI). The open symbols distinguish the BWR from the PWR samples. Different symbols are used for different data sets.

TABLE V

STATISTICAL CONTRIBUTION TO PRECISION IN ISOTOPIC FRACTION FOR 40-MIN COUNT

Plutonium Isotope	Average Precision (%) Obtained from Repeated Measurements
238	0.5
239	0.2
240	0.4
241	0.5
242	1.2

The isotopics data were analyzed as described in Sec. II.C.3 to give the results (relative to mass spectrometry) plotted in Figs. 12 through 16 vs the ^{240}Pu isotopic fraction. The empirical parameters used in this analysis are given in Table VI in the column labeled "original." (This table also references the equations in which the parameters are used.)

The "original" empirical parameters were derived based upon data that were limited in the type and number of reference samples. It is therefore appropriate to re-evaluate these parameters (using the complete 64-sample data set) during examination and evaluation of these techniques for determination of isotopic fractions.

The results for the individual isotopes are considered separately in the next five subsections followed by conclusions on the precision and accuracy of the technique for determination of isotopic fractions.

2. ^{242}Pu Results. The ^{242}Pu results plotted in Fig. 12 have a standard deviation of 25% (1σ) about the mean. The expected precision of the mass spectrometry results for ^{242}Pu, along with the contribution of the counting statistics of the passive assay, are an order of magnitude less. Therefore, the magnitude of the observed standard deviation is attributed primarily to the isotopic correlation. Although the PWR data lie relatively close to the zero line (indicating reasonable agreement with mass spectrometry), the BWR data

TABLE VI

ISOTOPICS PARAMETERS

<table>
<tr><th colspan="4">Empirical Parameters For Isotopics Analysis</th></tr>
<tr><th>Plutonium Isotope</th><th>Original</th><th>Updated</th><th>Updated With Modified Analysis</th></tr>
<tr><td>238
(low)</td><td>$K_{43} = 0.9840$</td><td>$K_{43} = 0.9840$</td><td>$K_{43} = 0.9840$</td></tr>
<tr><td></td><td colspan="2">(Equation 4)</td><td>(Equation 4)</td></tr>
<tr><td>240</td><td>$K_{45} = 1.0125$</td><td>$K_{45} = 1.0076$</td><td>$K_{45} = 1.0076$</td></tr>
<tr><td></td><td colspan="2">(Equation 5)</td><td>(Equation 5)</td></tr>
<tr><td>238
(high)</td><td>$a = 1.3236$</td><td>$a = 1.3236$</td><td>$a = 1.3236$</td></tr>
<tr><td></td><td colspan="2">(Equation 2)</td><td>(Equation 2)</td></tr>
<tr><td>241</td><td>$b = 1.2522$</td><td>$b = 1.2600$</td><td>$b_1 = 0.7606$
$b_2 = 0.1047$</td></tr>
<tr><td></td><td colspan="2">(Equation 3)</td><td>(Equation 9)</td></tr>
<tr><td>242</td><td>$C_1 = 0.5921$
$C_2 = -0.0547$</td><td>$\left.\begin{array}{l} C_1 = 0.7178 \\ C_2 = -0.0925 \end{array}\right\}$ PWR
$\left.\begin{array}{l} C_1 = 0.4955 \\ C_2 = -0.0468 \end{array}\right\}$ BWR</td><td>$\left.\begin{array}{l} C_1 = 0.7178 \\ C_2 = -0.0925 \end{array}\right\}$ PWR
$\left.\begin{array}{l} C_1 = 0.4955 \\ C_2 = -0.0468 \end{array}\right\}$ BWR</td></tr>
<tr><td></td><td colspan="2">(Equation 6)</td><td>(Equation 6)</td></tr>
</table>

show a positive deviation of 23%, on the average. This is because the isotopic correlation parameters, derived from a limited data set, are better suited to PWR than to BWR material.

The ^{242}Pu isotopic fractions for these process samples vary with burnup from 2 to 4%. Thus a 20% error in the ^{242}Pu isotopic ratio propagates through Eqs. (7) and (8) to nearly 1%, for the highest burnup samples, in the errors for the fractions of the other isotopes. Given the range and especially the extent of the burnup of the Tokai material, the use of a separate set of parameters for the ^{242}Pu isotopic correlation applied to BWR and PWR material causes a significant improvement in the results for the isotopic fractions. The mass spectrometry results for the fresh samples were fit to Eq. (6) to derive the separate parameters for the BWR and PWR material. These are given in Table VI in the "updated" column.

3. ^{241}Pu Results. The ^{241}Pu results plotted in Fig. 13 have a standard deviation of 1.5% (1σ) about the mean of -0.5%. Furthermore, a large (∿1%) systematic difference is observed between the average results for the BWR and PWR samples; this is due in large part to the systematics of the ^{242}Pu isotopic correlation. However, another effect is the apparent tendency of the results to vary with burnup. These effects are all large compared to the expected precision of the mass spectrometry and the contribution of the passive assay counting statistics.

Because the passive assay result for each plutonium isotopic fraction is correlated [through Eq. (7)] with that for each of the other isotopes, a more useful comparison (passive assay vs mass spectrometry) is that of the isotopic ratios (relative to ^{239}Pu), in order to independently deduce the magnitude of the biases and random effects. The ratio of ratios for ^{241}Pu $[(R_{241})_{\gamma} \div (R_{241})_{m.s.}]$ is plotted (vs the ^{240}Pu isotopic ratio) in Fig. 17. The standard deviation in the data (relative to the average result indicated by the dashed line) is 0.9% (1σ), an improvement over the 1.5% result, which includes the systematics of the ^{242}Pu isotopic correlation. However, a further improvement (to 0.7%) is achieved if the standard deviation is evaluated relative to a linear fit to the data in Fig. 17 (solid line).

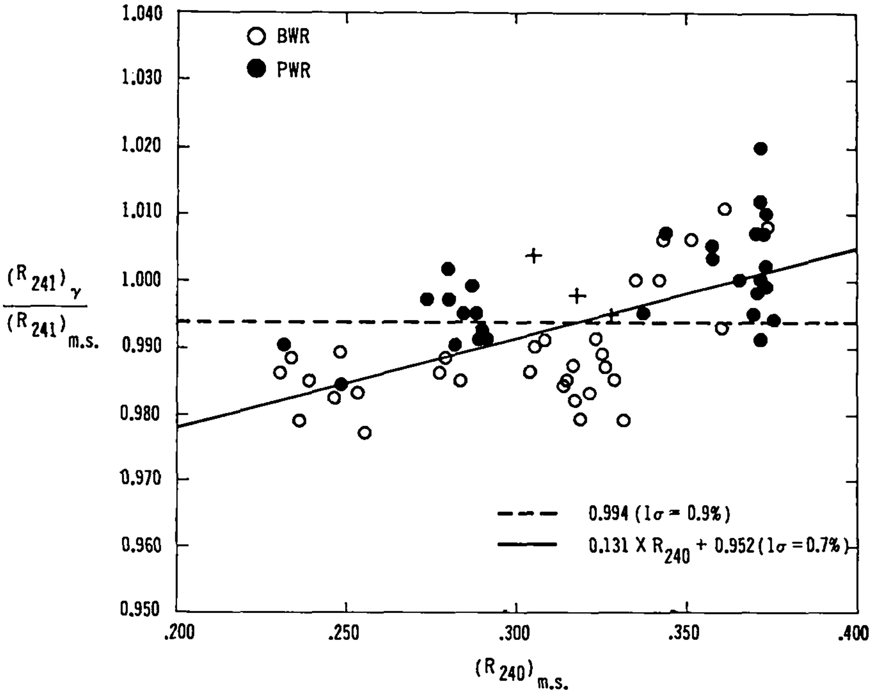

Fig. 17.
Ratio of the ^{241}Pu isotopic ratio (relative to ^{239}Pu) obtained from the passive gamma-ray assay to that obtained from mass spectrometry, plotted vs the ^{240}Pu isotopic ratio (relative to ^{239}Pu). The open symbols distinguish the BWR from the PWR samples. The dashed line corresponds to the average result. The solid line is the linear fit to the data. The 1-σ values are the percentage standard deviations of the data relative to the average result and relative to the linear fit.

The apparent change in the effective calibration parameter can only be due to one of three possible interferences: (1) the 277.6-keV gamma ray of ^{239}Np, which interferes with the ^{75}Se transmission gamma ray at 279.5 keV; (2) the pileup of the 59.6- and 88.0-keV gamma rays (of ^{241}Am and ^{109}Cd, respectively), which causes interference with the peak region of the 148.6-keV assay gamma ray of ^{241}Pu; or (3) the location of the low-energy background region for the 148.6-keV gamma ray between two ^{239}Pu peaks at 144.2 and 146.1 keV. Interferences (1) (refer to Sec. IV.A and Fig. 8) and (2) have been eliminated in that these two, even when combined, cannot account for as much as 0.1% of the (2%) effect observed over the range of burnup in Fig. 17. Furthermore, the February 1980 samples were assayed a second time after a 2-week delay. (Such a delay results in a disappearance of all evidence of ^{239}Np and an order-of-magnitude increase in ^{241}Am activity.) An average increase of 0.3% ± 0.3 (1σ) was observed in the ratio $(R_{241})_\gamma \div (R_{241})_{m.s.}$ obtained from the results of the second assay of these samples. This is sufficient to conclude that the interferences of ^{239}Np and ^{241}Am on the ^{241}Pu assay of the fresh samples are indeed negligible and that the ^{239}Pu gamma rays at 144.2- and 146.1-keV are the most likely causes of the changing ^{241}Pu bias.

An improvement in the absolute assay results for the ^{241}Pu isotopic fraction is achieved by adjusting the empirical parameter, b [Eq. (3)], by the factor $(0.994)^{-1}$. (Refer to the dashed line in Fig. 17.) An improvement in the standard deviation of these results relative to the mass spectrometry values is achieved by adjusting the original empirical parameter, b, by a quantity that is linear in R_{240}. A new empirical parameter, b', is thus defined using the empirical parameters for linear fit given in Fig. 17:

$$
\begin{aligned}
b' &= b \cdot (0.1311 \cdot R_{240} + 0.9524)^{-1} \\
&= (b_2 \cdot R_{240} + b_1)^{-1} \quad .
\end{aligned}
$$

Equation (3) can be rewritten with b' substituted for b:

$$
\frac{f_{241}}{f_{239}} = \frac{A(149)}{A(129)} \cdot \frac{CF(149)}{CF(129)} \cdot R_{\lambda MB} \left(\frac{241}{239}\right) \cdot b' = R_{241} \quad . \qquad (9)
$$

The original value of the parameter, b, and the adjusted (updated) value [that is, $b \cdot (0.994)^{-1}$] are given in Table VI. The parameters b_1 and b_2 appear in the column labeled "updated with modified analysis." "Modified analysis" refers to substitution of Eq. (9) for Eq. (3).

Several experimental observations reinforce the choice to use b' in Eq. (9) as described above. One is, of course, the improvement in the standard deviation of the ^{241}Pu results (relative to mass spectrometry) over the range of burnup observed at Tokai. The second observation is that the same linear dependence of the ^{241}Pu results on burnup (as defined in Fig. 17) has been shown for the (neptunium and americium-free) low-burnup samples ($0.066 < R_{240} < 0.193$) used for the preliminary calibration of this instrument at Los Alamos, indicating that Eq. (9) is valid over a much wider range of burnup than that represented by the Tokai samples. The third observation is that the results for $(R_{240})_\gamma \div (R_{240})_{m.s.}$ obtained with the fresh Tokai samples (refer to Fig. 18) are constant (within ±1%, 1σ) with burnup. The fourth observation is that the parameter b' is only slightly sensitive to R_{240}, such that a 1% uncertainty in R_{240} corresponds roughly to a 0.1% uncertainty in b'.

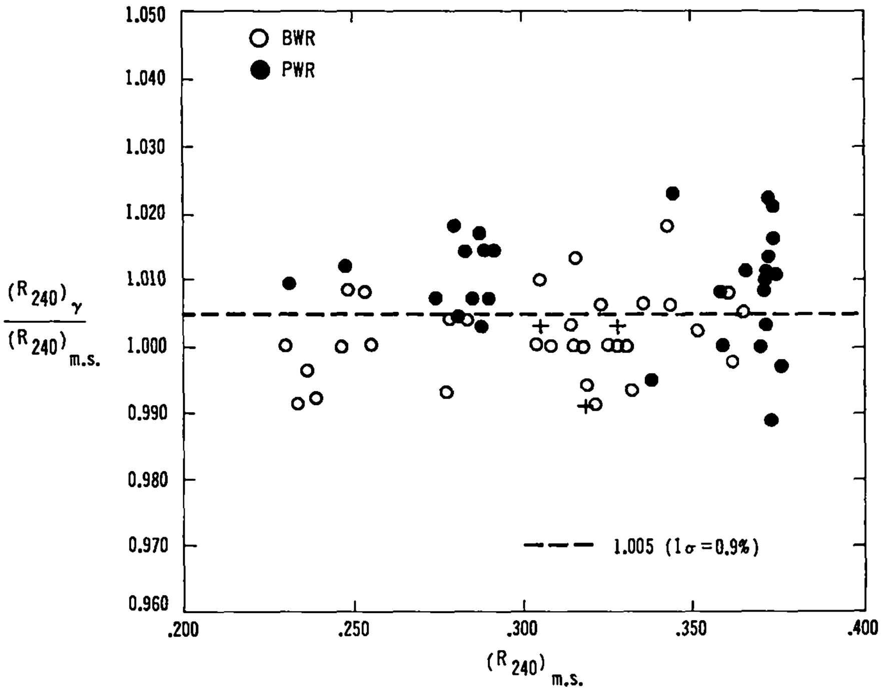

Fig. 18.
Ratio of the ^{240}Pu isotopic ratio (relative to ^{239}Pu) obtained from the passive gamma-ray assay to that obtained from mass spectrometry, plotted vs the ^{240}Pu isotopic ratio (relative to ^{239}Pu). The open symbols distinguish the BWR from the PWR samples. The dashed line corresponds to the average result. The 1-σ value is the percentage standard deviation of the data.

An option for this analysis is relocation of the low-energy background window for the 148.6-keV ^{241}Pu gamma-ray peak so that the ^{239}Pu interferences are removed. This is not practical because of the presence of an ^{241}Am gamma ray at 146.6 keV and the ^{241}Am-^{109}Cd pileup peak at 147.6 keV. Subtraction of such interferences in a consistent manner requires software of a complexity that would require substantial modification of the data analysis program.

4. ^{240}Pu Results. The ^{240}Pu results plotted in Fig. 14 have a standard deviation of 1.1% (1σ) about the mean of 0.4%. A systematic difference of 0.8% between the average results for the BWR and PWR samples is due primarily to the systematics of the ^{242}Pu isotopic correlation. The passive counting statistics as well as the expected precision in the mass spectrometry results represent only a small fraction of the standard deviation.

Figure 18 is a plot (analogous to Fig. 17) of the ratio of the ^{240}Pu isotopic ratios from passive assay and mass spectrometry vs the ^{240}Pu isotopic ratio. This plot shows the ^{240}Pu results independent of the correlated uncertainties that exist among the isotopic fractions. The magnitude of the bias in the empirical parameter is 0.5% with no evidence of dependence on burnup. A systematic difference of approximately 0.3% exists between the BWR and PWR samples. However, this difference is small relative to the standard deviation (0.9%, 1σ) in the data.

The empirical parameter, K_{45} [Eq. (5)], has been adjusted by the factor 1.005^{-1}. (Refer to the dashed line in Fig. 18.) The original and updated values of K_{45} are given in Table VI.

5. <u>^{239}Pu Results</u>. The ^{239}Pu results plotted in Fig. 15 scatter approximately ±0.5% (1σ) about the mean of -0.3%. Most of the negative bias is due to the BWR results, which on the average are systematically lower than those for the PWR samples by 0.3%. This systematic difference is primarily due to the systematics of the ^{242}Pu isotopic correlation.

The update of the six empirical calibration parameters including those for the ^{242}Pu isotopic correlation (refer to Table VI)--all determined from measured or derived isotopic ratios relative to ^{239}Pu--results in correlated changes that give a net improvement in the assay for ^{239}Pu. The results, given in Tables VII and VIII, are discussed in Sec. IV.E.7.

6. <u>^{238}Pu Results</u>. The ^{238}Pu results plotted in Fig. 16 show a scatter of ±15% (1σ) about the mean of -4%. Comparison of these results with independent passive assay results for the same samples using different analysis techniques (Task H of the TASTEX exercises[1]) gives relatively good agreement. Thus, most of the bias and scatter observed in Fig. 16 is attributed to mass-238 contamination affecting the mass spectrometry result, as discussed in Sec. III.B.2.

The ^{238}Pu isotopic fractions are typically 0.5%. Therefore, the magnitudes of the systematic and random effects introduced by the passive assay, although these cannot be determined quantitatively by the present evaluation, are too small (because the ^{238}Pu isotopic fraction is so small) to substantially affect the correlated results for the other isotopes.

7. <u>Precisions and Accuracies of Isotopic Fractions</u>. Tables VII and VIII summarize the results of analysis of the passive assay data using the three sets of empirical parameters given in Table VI. The updated parameters differ from the original parameters in that (1) the empirical constants, b (for ^{241}Pu) and K_{45} (for ^{240}Pu), have been optimized for the entire set of Tokai samples,

TABLE VII

ISOTOPICS RESULTS
PERCENT AVERAGE DEVIATION

Sample Type (N)	Isotopics Parameter Set	Plutonium Isotope				
		238	239	240	241	242
PWR (30)	Original	-3.19	-0.14	0.70	-0.23	0.81
	Updated	-3.31	-0.26	0.11	0.26	4.67
	Modified	-3.28	-0.23	0.12	0.12	4.62
BWR (34)	Original	-4.76	-0.39	-0.11	-1.42	23.12
	Updated	-4.33	0.08	-0.12	-0.34	3.47
	Modified	-4.33	0.08	-0.13	-0.17	3.43

TABLE VIII

ISOTOPICS RESULTS
PERCENT STANDARD DEVIATION

Sample Type (N)	Isotopics Parameter Set	Plutonium Isotope				
		238	239	240	241	242
PWR (30)	Original	2.77	0.33	0.63	0.78	8.51
	Updated	2.71	0.34	0.64	0.69	8.07
	Modified	2.72	0.33	0.63	0.61	8.01
BWR (34)	Original	15.56	0.44	0.97	0.81	13.88
	Updated	15.48	0.39	0.99	0.87	11.62
	Modified	15.47	0.38	0.99	0.76	11.62

and (2) separate sets of parameters for the ^{242}Pu isotopic correlation have been derived for BWR and PWR samples. The modified results were obtained with the same parameters as those used to obtain the updated results except that the parameter for the ^{241}Pu analysis includes a burnup-dependence.

Table VII lists the average percentage differences between the passive analysis and the mass spectrometry results. Table VIII lists the standard deviation (1σ) in the relative results. (The relative results are the percentage differences between the passive analysis and mass spectrometry results.)

It is assumed that the mass spectrometry results are the true values for the isotopic fractions of ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{242}Pu. Therefore, the average deviations give the inherent bias in the passive assay techniques applied to the fresh Tokai samples. Furthermore, because the expected precisions in the mass spectrometry results are significantly better for ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{242}Pu than the observed standard deviations, the standard deviations can be used to determine the accuracy limits for the passive assay technique.

In general, significant improvements are observed in the average deviations for ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{242}Pu in the updated analysis results. Further improvement in this quantity is achieved for ^{241}Pu with the modified analysis. The existence of a net bias in the assay results is due to the small systematic differences in the K_{45} and b parameters (for ^{240}Pu and ^{241}Pu analysis, respectively) observed in the BWR and PWR results of Figs. 17 and 18. These differences cause the average bias for ^{240}Pu and ^{241}Pu to be slightly positive for the PWR data and slightly negative for the BWR data. This occurs because the average result for each of these (b and K_{45}) parameters is used to analyze all data. The (correlated) results for the other isotopes are also affected. However, the average deviations are smaller than the standard deviations (by a factor that varies between 2 and 10); that is, the bias in the passive assay result is small compared to the accuracy for ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{242}Pu.

The precisions for the isotopic fractions measured in repeated, 2000-second assays (Table VI) give the contributions of counting statistics to the overall accuracy of a 2000-second assay. The standard deviation values given in Table VIII do not reflect this contribution because the results shown in

Table VIII are obtained from the average result of 10 complete passive assay cycles. The accuracies of the isotopic fractions obtained in a single-assay cycle (which corresponds to a 2000-second passive count) can be obtained from the quadratic sum of the standard deviation (Table VIII) and the precision (Table VI). This assumes that the precisions in the mass spectrometry results do not contribute significantly to the relative isotopics results. The accuracy computed for each isotope in this way is given separately for the PWR and BWR samples in Table IX. The results were computed using the standard deviations obtained with the modified parameter set, but the results are substantially the same, regardless of the choice of parameters. If the contributions of the precision in the mass spectrometry (as given in Sec. III.B.2) are unfolded from these quantities, the computed accuracies for the ^{240}Pu, ^{241}Pu, and ^{242}Pu isotopic fractions obtained from a 2000-second passive assay are each improved by 0.1% over the values in Table IX.

Appendix E contains tabulations of the reference values and the passive gamma-ray results for the isotopic weight ratios. Separate tabulations are given for PWR and BWR samples. The passive gamma-ray results are computed using both the original and the modified parameter sets.

TABLE IX

ACCURACY OF 2000-SECOND ISOTOPICS ASSAY

Sample Type	Plutonium Isotope				
	238	239	240	241	242
PWR (30 samples)	—	0.4%	0.7%	0.8%	8.2%
BWR (34 samples)	—	0.4%	1.1%	0.9%	11.7%

V. CONCLUSIONS AND RECOMMENDATIONS

A. Plutonium Concentration Assay

The K-edge results for plutonium concentration assay have been evaluated relative to those of destructive chemical analysis performed on the same product solution samples. The following concluding statements can be made concerning the K-edge measurements.

1. The overall accuracy of the K-edge technique for plutonium concentration assay, determined during a 1-year period of measurements of process samples, is 0.6% or better. This estimate assumes that (a) the reference concentrations obtained by destructive analysis of the same process samples do not contribute to the uncertainty in the relative results, and (b) other effects (caused by sampling or evaporation, for example) that might lead to systematic differences between the K-edge and reference results are not significant.

2. A systematic difference of approximately 0.1% between the concentration assays for aged and fresh samples is observed (at the 70% confidence level) in these applications.

3. The precision, including the effects of sample positioning, obtained in 40-minute count periods, is approximately 0.2%. This result, which agrees with the calculated contribution of counting statistics, indicates that random effects associated with routine sample positioning can be ignored.

4. The relative results for plutonium concentration (K-edge relative to destructive chemical analysis) indicate that the calibration for the K-edge assay is biased negatively by approximately 0.3%.

5. The measurement control data obtained by routine K-edge assays of a plutonium foil during the 1-year period indicate that random effects (other than counting statistics) contribute 0.14% or less to the K-edge uncertainty.

Anomalously large systematic effects, such as the low average of the relative results for the October 1980 samples, have added significantly to the limits in the uncertainty (item 1 above) determined for the K-edge measurements in this evaluation at Tokai. It would be useful to incorporate measurement control procedures that apply simultaneously to the K-edge and the destructive assay methods (such as measurements of primary standard plutonium samples or measurements by a third, independent technique) to attempt to isolate the origin of such systematic effects in the relative results.

B. Plutonium Isotopics Assay

The assay of the isotopic fractions of ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{242}Pu based upon passive gamma-ray counting with self-attenuation corrections derived from measured transmissions has been evaluated (by comparisons with mass spectrometry) for measurements of fresh product solutions. The following conclusions can be made concerning these measurements.

1. The bias in the assay of individual isotopes is greatly reduced if a separate set of parameters is used for the ^{242}Pu isotopic correlation for BWR and PWR samples.

2. The precisions in the isotopic fractions determined from a 2000-second passive count are 0.2% for ^{239}Pu, 0.4% for ^{240}Pu, 0.5% for ^{241}Pu, and 1.2% for ^{242}Pu.

3. The accuracies (including counting statistics) of the passive assay determined from a 2000-second passive count (for the range of samples assayed) are 0.4% for ^{239}Pu, 1.0% for ^{240}Pu, 0.8% for ^{241}Pu, and 10% for ^{242}Pu, using separate isotopic correlations for BWR and PWR samples.

4. The average net bias for each isotope is smaller than the corresponding accuracy given in item 3 above by a factor that varies between 2 and 10.

The comparison of the passive assay results for the ^{238}Pu isotopic fraction with ^{238}Pu determinations by alpha counting techniques would enable evaluation of the passive gamma techniques for determination of this isotope.

C. Recommendations and Extended Applications

1. The K-Edge Measurement Technique. The inherent stability of the K-edge calibration, the relatively low sensitivity of the K-edge technique to changes in sample matrix and radioactivity, and the relatively small contribution of random effects to the overall uncertainty in the K-edge results are all advantages to reprocessing applications where samples are likely to vary in composition and where the process environment requires timely, reliable measurements without the need for frequent recalibration. Some improvements on the existing Tokai instrument would be accomplished with additional attenuation of the 59.6-keV gamma rays from ^{241}Am, which currently introduce a small systematic shift in the assay results for very aged samples. This attenuation might be accomplished with absorber foils inserted in the collimators for the transmission sources.

The availability of three additional positions in the source wheel of the mechanical system leaves open the possibility for modification of the Tokai instrument for uranium solution assay. Measurement techniques that are currently being applied in an existing instrument[10] to give uranium concentration for materials accounting and process control could also be incorporated, as additional capabilities, into the Tokai equipment.

The adaptation of this K-edge technique to on-line operation is currently being demonstrated at the Savannah River Plant with measurements of precipitator feed solutions in a bypass line in the reprocessing facility.[11,12] The use of automated computer-based absorption-edge instruments to provide on-line measurement of special-nuclear-material (SNM) concentration in near-real-time for materials accounting has recently been demonstrated[13] with an L-edge device. Similar applications of the K-edge technique used in the Tokai instrument might be considered.

Measurements of uranium and plutonium concentration in fast-breeder reactor dissolver solutions have recently been demonstrated using the K-edge technique, intense continuum sources, and highly restrictive sample collimation.[14] The K-edge technique used in the Tokai instrument might be similarly applied to process samples beyond the first extraction[15] where fission product levels are too high to be compatible with other types of nondestructive assay.

The simplicity of the K-edge technique used in the Tokai instrument makes the method suitable for incorporation into portable instrumentation equipped with compact, programmable analysis capabilities.[16] With the advent of compact, rugged, low-maintenance detectors with intermediate resolution (such as the CdTe detector), the methods might also extend to in-line applications in remote maintenance areas.

2. The Passive Gamma-Ray Assay Technique. Use of ratios of areas of closely spaced gamma-ray peaks to determine the isotopics of plutonium samples is a technique that is quite insensitive to changes in detector efficiency, electronic gain, and counting geometry. Thus, the passive assay measurements are useful in plant environments because frequent recalibration is not required.

This technique does not require the fitting of spectral data to obtain precise and accurate results for the plutonium isotopic fractions. The analysis software is sufficiently simple so that only the minimum memory and computing capabilities of commercial computer-based MCAs are required. The technique can even be accommodated, in principle, by the limited memory capabilities of some of the new, portable MCAs that are equipped for acquisition of high-resolution gamma-ray data.[16] In all cases, the assay results are timely.

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APPENDIX A

COMPUTATION OF CORRECTION FACTORS FOR SELF-ATTENUATION OF 129-, 149-, AND 153-keV GAMMA RAYS.

The correction factors for self-attenuation of the 129-, 149-, and 153-keV passive gamma rays take the form (for slab absorbers):

$$CF = D \div \left[X_0 \cdot (X_0 + D) \cdot DI \cdot \sum_{I=1}^{N} \frac{e^{-\mu \cdot (I-0.5) \cdot DI}}{(X_0 + (I-0.5) \cdot DI)^2} \right] ,$$

where D is the sample thickness, DI is the thickness of each thin slab of sample over which the numerical integration is carried out, N is the number of slabs in the sample, X_0 is the detector-to-sample distance, and μ is the empirically determined sample attenuation coefficient.

The μ values are determined from the expression

$$T = e^{-\mu x} ,$$

where the transmission values, T, are obtained for each gamma-ray assay peak energy by extrapolation (or interpolation) of the measured transmissions at 122.1, 136.0, and 279.5 keV (E_1, E_2, and E_3, respectively). A linear extrapolation (or interpolation) is performed on the quantities $\ell n(-\ell n T_I)$ vs $\ell n E_I$ based on data obtained at energies E_1 and E_2. A separate interpolation (or extrapolation) is performed based on the data obtained at E_2 and E_3. Thus, two transmission values, T_A and T_B, for each energy E (corresponding to 129.3, 148.6, or 152.7 keV) are obtained. The transmission, T, at each energy, E, is the average of T_A and T_B. Analytically, this is

$$\ell n(-\ell n T_A) = a_A + b_A \cdot \ell n E$$

$$\ell n(-\ell n T_B) = a_B + b_B \cdot \ell n E$$

$$\ell n(-\ell n T) = (a_A + a_B + (b_A + b_B) \cdot \ell n E) \div 2 = Y \quad ,$$

where

$$T = e^{(-e^Y)} \; .$$

APPENDIX B

CONSTANTS USED IN EVALUATION OF ISOTOPIC WEIGHT RATIOS USING THE HIGH-ENERGY PASSIVE DATA

$$R_{\lambda M B}\left(\frac{238}{239}\right) = \frac{\lambda_{239} \cdot B_{129} \cdot M_{238}}{\lambda_{238} \cdot B_{153} \cdot M_{239}} = \frac{2.8783 \times 10^{-5}}{7.8955 \times 10^{-3}} \cdot \frac{6.26 \times 10^{-5}}{9.56 \times 10^{-6}} \cdot \frac{238.05}{239.05}$$

$$= 0.023771$$

$$R_{\lambda M B}\left(\frac{241}{239}\right) = \frac{\lambda_{239} \cdot B_{129} \cdot M_{241}}{\lambda_{241} \cdot B_{149} \cdot M_{239}} = \frac{2.8783 \times 10^{-5}}{4.8303 \times 10^{-2}} \cdot \frac{6.26 \times 10^{-5}}{1.87 \times 10^{-6}} \cdot \frac{241.06}{239.05}$$

$$= 0.0201155$$

APPENDIX C

CONSTANTS USED IN EVALUATION OF ISOTOPIC WEIGHT RATIOS USING THE LOW-ENERGY PASSIVE DATA

$$R_{\lambda MB}\left(\frac{43}{52}\right) = \frac{\lambda_{239} \cdot B_{52} \cdot M_{238}}{\lambda_{238} \cdot B_{43} \cdot M_{239}} = \frac{87.79}{24082} \cdot \frac{2.7 \times 10^{-4}}{3.93 \times 10^{-4}} \cdot \frac{238.05}{239.05}$$

$$= 0.002494$$

$$R_{\lambda MB}\left(\frac{45}{52}\right) = \frac{\lambda_{239} \cdot B_{52} \cdot M_{240}}{\lambda_{240} \cdot B_{45} \cdot M_{239}} = \frac{6537}{24082} \cdot \frac{2.7 \times 10^{-4}}{4.53 \times 10^{-4}} \cdot \frac{240.03}{239.05}$$

$$= 0.16245$$

APPENDIX D

TABULATION OF CONCENTRATION RESULTS

Sample Set	No.	Sample ID	F or A	ρ_K(g/ℓ)	δ(%)	ρ_{REF}(g/ℓ)	$\frac{\rho_K - \rho_{REF}}{\rho_{REF}} \times 100$
Oct. '79 (calib.)	1	F3	F	142.7	n.a.	144.0	-0.92
	2	F4	F	170.7	n.a.	171.4	-0.41
	3	F5	F	197.9	n.a.	197.3	0.30
	4	F6	F	222.9	n.a.	221.1	0.82
	5	NF3	F	185.9	n.a.	186.6	-0.40
	6	NF4	F	215.9	n.a.	216.6	-0.31
	7	NF6	F	253.5	n.a.	253.8	-0.13
	8	NF7	F	341.0	n.a.	338.5	0.73
	9	A3	A	175.0	n.a.	175.3	-0.15
	10	A4	A	204.3	n.a.	205.0	-0.33
	11	A5	A	231.6	n.a.	231.5	0.05
	12	A6	A	270.0	n.a.	268.0	0.75
Nov. '79 (camp.)	13	PG1	F	213.1	0.58	212.1	0.47
	14	PG2	F	258.7	0.12	255.9	1.09
	15	PG3	F	231.5	0.25	231.2	0.13
	16	PG4	F	272.3	0.29	269.6	1.02
	17	PG5	F	254.1	0.21	254.3	-0.08
	18	PG6	A	216.9	n.a.	217.1	-0.09
	19	PG7	A	246.5	n.a.	244.4	0.86
Jan. '80 (camp.)	20	PG8	F	206.4	0.17	203.2	1.57
	21	PG9	F	232.6	0.00	230.1	1.09
	22	PG10	F	239.9	0.00	237.9	0.83
	23	PG11	F	245.9	0.17	245.5	0.16
	24	PG12	F	242.8	0.26	240.1	1.12
	25	PG13	F	234.3	0.12	233.5	0.34
	26	PG14	F	224.3	0.06	223.1	0.54
March '80 (inv., trans.)	27	PG15	A	253.5	0.35	253.8	-0.11
	28	PG16	A	238.3	0.45	240.4	-0.87
	29	PG17	A	240.3	0.25	241.0	-0.27

Sample Set	No.	Sample ID	F or A	ρ_K(g/ℓ)	δ(%)	ρ_{REF}(g/ℓ)	$\frac{\rho_K-\rho_{REF}}{\rho_{REF}}$x 100
	30	PG18	A	241.5	0.00	243.1	-0.65
	31	PG19	A	234.5	0.46	235.8	-0.57
	32	PG20	A	235.5	0.18	236.9	-0.57
	33	PG21	A	240.0	0.15	242.0	-0.82
May '80	34	PG22	F	181.5	0.63	184.1	-1.39
(camp.)	35	PG23	F	214.9	0.50	216.5	-0.74
	36	PG24	A	220.7	0.71	221.3	-0.03
	37	PG25	F	222.0	0.29	220.3	0.75
	38	PG26	F	218.7	0.46	219.1	-0.18
	39	PG27	A	220.9	0.13	224.4	-1.60
	40	PG28	F	214.9	0.20	215.6	-0.32
	41	PG29	F	168.1	0.59	169.4	-0.75
	42	PG30	A	222.8	0.06	224.0	-0.55
April '80	43	PG31	F	181.5	n.a.	181.1	0.22
(demo.)	44	PG32	F	219.4	n.a.	220.0	-0.27
	45	PG33	F	253.8	n.a.	253.6	0.08
	46	PG34	A	154.9	n.a.	155.3	-0.26
	47	PG35	A	195.7	n.a.	196.4	-0.36
	48	PG36	A	209.8	n.a.	211.7	-0.90
	49	PG37	A	243.6	n.a.	243.5	0.04
	50	PG38	U[a]	234.0	n.a.	233.7	0.13
	51	PG39	U[a]	238.6	n.a.	238.0	0.25
June '80	52	PG40	A	225.4	0.04	225.6	-0.09
(camp.)	53	PG41	F	183.0	0.22	185.1	-1.13
	54	PG42	A	228.6	0.41	229.5	-0.39
	55	PG43	F	158.3	0.06	160.1	-1.12
	56	PG44	F	182.7	0.16	183.8	-0.60
	57	PG45	F	179.8	0.16	182.1	-1.26
	58	PG46	F	202.0	0.20	202.8	-0.39
	59	PG47	A	233.1	0.12	236.0	-1.23

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[a] U = unknown.

Sample Set	No.	Sample ID	F or A	ρ_K(g/ℓ)	δ(%)	ρ_{REF}(g/ℓ)	$\frac{\rho_K-\rho_{REF}}{\rho_{REF}}$ x 100
	60	PG48	F	190.8	0.25	191.8	-0.52
	61	PG49	F	215.2	0.07	217.4	-1.01
	62	PG50	F	227.8	0.01	227.6	0.09
	63	PG51	F	214.5	0.43	215.7	-0.57
	64	PG52	A	239.7	0.24	239.6	0.04
	65	PG53	F	221.7	0.34	221.7	0.00
	66	PG54	F	211.3	0.06	210.6	0.33
	67	PG55	F	207.1	0.18	208.1	-0.48
	68	PG56	F	211.7	0.00	210.3	0.67
	69	PG57	F	219.6	0.74	219.1	0.23
	70	PG58	F	246.2	0.32	247.3	-0.44
	71	PG59	F	228.4	0.84	226.9	0.66
	72	PG60	F	221.9	0.41	222.5	-0.27
	73	PG61	A	215.3	0.70	215.3	0.00
	74	PG62	F	153.5	0.63	153.8	-0.20
Sept. '80	75	PG63	A	216.0	0.26	219.7	-1.68
(camp.)	76	PG64	A	221.6	0.61	220.7	0.41
	77	PG65	A	219.7	0.26	224.0	-2.10
	78	PG66	A	221.0	0.26	221.8	-0.36
	79	PG67	A	223.2	0.06	224.2	-0.45
	80	PG68	A	223.8	0.28	225.8	-0.89
	81	PG69	A	210.7	0.10	215.1	-2.05
	82	PG70	A	211.6	0.13	216.2	-2.13
	83	PG71	A	211.4	0.13	213.3	-0.89
	84	PG72	A	213.8	0.17	214.7	-0.42
	85	PG73	F	177.2	0.20	177.1	0.06
	86	PG74	F	230.0	0.34	230.2	-0.09
	87	PG75	F	219.2	0.16	218.9	0.14
	88	PG76	F	190.5	0.22	192.1	-0.83
	89	PG77	A	216.5	0.13	218.9	-1.10
	90	PG78	F	231.7	0.98	232.7	-0.43

Sample Set	No.	Sample ID	F or A	ρ_K(g/ℓ)	δ(%)	ρ_{REF}(g/ℓ)	$\frac{\rho_K-\rho_{REF}}{\rho_{REF}} \times 100$
	91	PG79	F	212.6	0.20	213.5	-0.42
	92	PG80	F	220.8	0.10	220.3	0.23
	93	PG81	F	261.5	0.05	259.8	0.65
	94	PG82	F	129.8	0.27	133.2	-2.55
Oct. '80	95	PG83	A	220.5	0.23	223.2	-1.21
(camp.)	96	PG84	A	222.3	0.04	224.4	-0.94
	97	PG85	A	224.2	0.12	225.2	-0.44
	98	PG86	F	160.7	0.22	163.5	-1.71
	99	PG87	F	218.9	0.08	220.0	-0.50
	100	PG88	F	154.8	0.34	159.4	-2.89
	101	PG89	F	151.9	0.41	153.2	-0.85
	102	PG90	F	229.8	0.34	233.5	-1.59
	103	PG91	F	228.7	0.14	231.0	-1.00
	104	PG92	F	230.4	0.16	235.3	-2.08
	105	PG93	F	218.2	0.06	220.5	-1.04
	106	PG94	F	194.5	0.18	196.5	-1.02
	107	PG95	A	232.4	0.17	235.4	-1.27
	108	PG96	F	211.9	0.37	213.5	-0.75
	109	PG97	F	195.9	0.13	198.6	-1.36
	110	PG98	A	234.2	0.05	236.4	-0.93
	111	PG99	F	219.1	0.18	219.8	-0.32
	112	PG100	A	238.4	0.04	237.9	0.21
Jan. '81	113	PG101	A	241.8	0.06	242.0	-0.10
(camp.)	114	PG102	A	243.1	0.13	246.8	-1.52
	115	PG103	A	233.6	0.04	235.3	-0.74
	116	PG104	A	235.9	0.13	238.1	-0.90
	117	PG105	A	239.0	0.00	241.3	-0.97
	118	PG106	A	241.5	0.00	241.6	-0.04
	119	PG107	A	221.4	0.05	221.9	-0.25
	120	PG108	F	162.2	0.08	162.9	-0.45
	121	PG109	F	174.9	0.22	174.9	0.01

Sample Set	No.	Sample ID	F or A	$\rho_K(g/\ell)$	$\delta(\%)$	$\rho_{REF}(g/\ell)$	$\frac{\rho_K-\rho_{REF}}{\rho_{REF}} \times 100$
	122	PG110	A	231.2	0.16	231.7	-0.22
	123	PG111	A	222.8	0.19	223.7	-0.44
	124	PG112	F	206.5	0.09	209.0	-1.22
	125	PG113	A	222.1	0.22	224.1	-0.86
	126	PG114	F	177.9	0.12	177.7	0.11
	127	PG115	A	224.9	0.20	224.1	0.33
	128	PG116	A	224.6	0.09	223.2	0.63
	129	PG117	A	225.5	0.22	224.7	0.34
	130	PG118	F	191.8	0.03	190.6	0.61
	131	PG119	F	217.3	0.05	215.7	0.72
	132	PG120	A	227.3	0.20	225.4	0.87
	133	PG121	A	230.0	0.20	227.5	1.11

APPENDIX E

TABULATION OF ISOTOPIC WEIGHT RATIO RESULTS

TABLE E-1

ISOTOPIC WEIGHT RATIOS FOR PWR SAMPLES: PASSIVE γ RESULTS COMPUTED WITH ORIGINAL PARAMETERS

No.	Sample ID		%238	%239	%240	%241	%242
1	4	REF	0.836	65.121	22.023	8.806	3.215
		γ	0.823	65.320	21.920	8.790	3.160
		% DEV	-1.555	0.306	-0.468	-0.182	-1.711
2	5	REF	1.109	62.975	22.552	9.658	3.706
		γ	1.100	62.930	22.690	9.680	3.600
		% DEV	-0.812	-0.071	0.612	0.228	-2.860
3	8	REF	0.990	63.286	22.679	9.384	3.661
		γ	0.991	63.320	22.680	9.440	3.570
		% DEV	0.101	0.054	0.004	0.597	-2.486
4	9	REF	0.982	62.490	23.135	9.568	3.825
		γ	0.971	62.610	23.090	9.540	3.780
		% DEV	-1.120	0.192	-0.195	-0.293	-1.176
5	10	REF	1.009	62.383	23.159	9.593	3.856
		γ	0.982	62.210	23.280	9.640	3.890
		% DEV	-2.676	-0.277	0.522	0.490	0.882
6	11	REF	0.995	62.270	23.192	9.670	3.873
		γ	0.973	62.120	23.340	9.650	3.920
		% DEV	-2.211	-0.241	0.638	-0.207	1.214

No.	Sample ID		%238	%239	%240	%241	%242
7	12	REF	0.979	62.269	23.269	9.612	3.871
		γ	0.976	61.780	23.580	9.630	4.040
		% DEV	-0.306	-0.785	1.337	0.187	4.366
8	13	REF	1.019	62.208	23.173	9.728	3.872
		γ	1.016	62.410	23.030	9.780	3.770
		% DEV	-0.294	0.325	-0.617	0.535	-2.634
9	14	REF	1.169	61.636	23.166	10.050	3.978
		γ	1.163	61.770	23.170	10.020	3.880
		% DEV	-0.513	0.217	0.017	-0.299	-2.464
10	22	REF	1.097	66.963	20.403	8.219	3.318
		γ	1.051	67.390	20.640	8.300	2.620
		% DEV	-4.193	0.638	1.162	0.986	-21.037
11	73	REF	1.019	63.840	21.960	9.834	3.347
		γ	0.977	63.430	22.320	9.840	3.430
		% DEV	-4.122	-0.642	1.639	0.061	2.480
12	74	REF	1.247	61.302	22.431	11.033	3.987
		ρ	1.236	61.290	22.700	11.030	3.740
		% DEV	-0.882	-0.020	1.199	-0.027	-6.195
13	75	REF	1.416	60.526	22.528	11.376	4.154
		γ	1.316	60.340	22.950	11.480	3.910
		% DEV	-7.062	-0.307	1.873	0.914	-5.874
14	76	REF	1.331	60.638	22.524	11.604	4.102
		γ	1.293	60.760	22.600	11.600	3.740
		% DEV	-2.855	0.201	0.337	-0.034	-8.825

No.	Sample ID		%238	%239	%240	%241	%242
15	79	REF	1.339	60.493	22.565	11.452	4.150
		γ	1.320	60.390	22.890	11.510	3.890
		% DEV	-1.419	-0.170	1.440	0.506	-6.265
16	80	REF	1.324	60.619	22.529	11.401	4.127
		γ	1.308	60.480	22.780	11.600	3.840
		% DEV	-1.208	-0.229	1.114	1.745	-6.954
17	81	REF	1.374	60.523	22.542	11.416	4.145
		γ	1.312	60.570	22.850	11.410	3.850
		% DEV	-4.512	0.078	1.366	-0.053	-7.117
18	82	REF	1.382	60.614	22.560	11.319	4.125
		γ	1.278	60.770	22.860	11.250	3.840
		% DEV	-7.525	0.257	1.330	-0.610	-6.909
19	86	REF	0.628	74.051	17.132	6.575	1.614
		γ	0.615	74.165	17.264	6.520	1.435
		% DEV	-2.070	0.154	0.770	-0.836	-11.090
20	87	REF	0.564	73.228	18.146	6.405	1.657
		γ	0.524	73.118	18.342	6.294	1.721
		% DEV	-7.092	-0.150	1.080	-1.733	3.862
21	88	REF	0.584	70.617	19.883	6.836	2.081
		γ	0.566	70.305	20.095	6.733	2.302
		% DEV	-3.082	-0.442	1.066	-1.507	10.620
22	89	REF	0.593	69.901	20.328	6.968	2.210
		γ	0.577	69.586	20.504	6.876	2.456
		% DEV	-2.698	-0.451	0.866	-1.320	11.131

No.	Sample ID		%238	%239	%240	%241	%242
23	90	REF	0.595	69.999	20.257	6.957	2.192
		γ	0.575	69.953	20.229	6.885	2.327
		% DEV	-3.361	-0.066	-0.138	-1.035	6.159
24	91	REF	0.594	69.990	20.295	6.935	2.186
		γ	0.574	69.777	20.378	6.861	2.409
		% DEV	-3.367	-0.304	0.409	-1.067	10.201
25	92	REF	0.555	70.846	19.860	6.714	2.025
		γ	0.539	70.438	20.077	6.655	2.290
		% DEV	-2.883	-0.576	1.093	-0.879	13.086
26	93	REF	0.577	70.226	20.169	6.883	2.145
		γ	0.569	69.775	20.405	6.834	2.417
		% DEV	-1.386	-0.642	1.170	-0.712	12.681
27	94	REF	0.642	70.339	20.056	6.837	2.126
		γ	0.558	70.176	20.156	6.785	2.325
		% DEV	-13.084	-0.232	0.499	-0.761	9.360
28	96	REF	0.598	70.152	20.182	6.909	2.159
		γ	0.570	70.001	20.216	6.862	2.351
		% DEV	-4.682	-0.215	0.168	-0.680	8.893
29	97	REF	0.551	70.944	19.842	6.661	2.002
		γ	0.526	70.699	19.897	6.650	2.227
		% DEV	-4.537	-0.345	0.277	-0.165	11.239
30	99	REF	0.537	71.431	19.551	6.569	1.912
		γ	0.514	71.164	19.655	6.528	2.139
		% DEV	-4.283	-0.374	0.532	-0.624	11.872
AV DEV (%)			-3.1898	-0.1373	0.7035	-0.2258	0.8149
ST DEV (%)			2.7650	0.3337	0.6355	0.7844	8.5119

TABLE E-2

ISOTOPIC WEIGHT RATIOS FOR BWR SAMPLES: PASSIVE γ RESULTS COMPUTED WITH ORIGINAL PARAMETERS

No.	Sample ID		%238	%239	%240	%241	%242
1	1	REF	0.347	74.555	18.368	5.445	1.285
		γ	0.332	74.380	18.290	5.330	1.660
		% DEV	-4.323	-0.235	-0.425	-2.112	29.183
2	2	REF	0.517	71.161	19.695	6.668	1.960
		γ	0.495	71.260	19.560	6.580	2.110
		% DEV	-4.255	0.139	-0.685	-1.320	7.653
3	3	REF	0.734	66.953	21.299	8.204	2.810
		γ	0.716	67.110	21.170	8.210	2.800
		% DEV	-2.452	0.234	-0.606	0.073	-0.356
4	22	REF	1.097	66.963	20.403	8.219	3.318
		γ	1.051	67.390	20.640	8.300	2.620
		% DEV	-4.193	0.638	1.162	0.986	-21.037
5	23	REF	0.720	70.349	19.638	6.956	2.337
		γ	0.698	70.480	19.740	6.890	2.200
		% DEV	-3.056	0.186	0.519	-0.949	-5.862
6	25	REF	0.444	73.656	18.657	5.747	1.496
		γ	0.414	73.450	18.700	5.640	1.800
		% DEV	-6.757	-0.280	0.230	-1.862	20.321
7	26	REF	0.445	74.105	18.385	5.799	1.266
		γ	0.419	73.810	18.380	5.680	1.710
		% DEV	-5.843	-0.398	-0.027	-2.052	35.071

No.	Sample ID		%238	%239	%240	%241	%242
8	28	REF	0.451	74.764	17.841	5.836	1.108
		γ	0.794	74.660	17.690	5.740	1.510
		% DEV	76.053	-0.139	-0.846	-1.645	36.282
9	29	REF	0.465	74.640	17.640	6.134	1.121
		γ	0.415	74.550	17.550	6.000	1.490
		% DEV	-10.753	-0.121	-0.510	-2.185	32.917
10	41	REF	0.544	74.816	17.409	6.074	1.157
		γ	0.422	74.840	17.320	6.000	1.430
		% DEV	-22.426	0.032	-0.511	-1.218	23.596
11	43	REF	0.397	75.265	17.303	5.979	1.056
		γ	0.402	75.050	17.260	5.880	1.410
		% DEV	1.259	-0.286	-0.249	-1.656	33.523
12	44	REF	0.575	72.696	18.526	6.703	1.500
		γ	0.548	72.560	18.560	6.540	1.800
		% DEV	-4.696	-0.187	0.184	-2.432	20.000
13	45	REF	0.584	70.369	18.882	7.289	1.876
		γ	0.541	70.340	19.740	7.180	2.200
		% DEV	-7.363	-0.041	4.544	-1.495	17.271
14	46	REF	0.620	68.304	21.044	7.808	2.224
		γ	0.569	68.100	20.950	7.720	2.670
		% DEV	-8.226	-0.299	-0.447	-1.127	20.054
15	48	REF	0.627	67.150	21.718	8.086	2.419
		γ	0.593	66.730	21.710	7.960	2.990
		% DEV	-5.423	-0.625	-0.037	-1.558	23.605

No.	Sample ID		%238	%239	%240	%241	%242
16	49	REF	0.621	67.324	21.648	8.027	2.380
		γ	0.583	67.300	21.430	7.890	2.870
		% DEV	-6.119	-0.036	-1.007	-1.707	20.588
17	50	REF	0.606	67.618	21.488	7.953	2.335
		γ	0.563	67.530	21.320	7.770	2.820
		% DEV	-7.096	-0.130	-0.782	-2.301	20.771
18	51	REF	0.626	67.653	21.443	7.946	2.332
		γ	0.568	67.460	21.350	7.790	2.830
		% DEV	-9.265	-0.285	-0.434	-1.963	21.355
19	53	REF	0.649	67.006	21.797	8.107	2.441
		γ	0.596	66.760	21.670	7.990	2.980
		% DEV	-8.166	-0.367	-0.583	-1.443	22.0810
20	54	REF	0.662	66.692	21.951	8.200	2.495
		γ	0.613	66.410	21.860	8.050	3.070
		% DEV	-7.402	-0.423	-0.415	-1.829	23.046
21	55	REF	0.650	66.591	22.008	8.243	2.508
		γ	0.603	66.520	21.780	8.060	3.030
		% DEV	-7.231	-0.107	-1.036	-2.220	20.813
22	56	REF	0.625	66.965	21.831	8.131	2.448
		γ	0.601	66.660	21.690	7.990	3.010
		% DEV	-3.840	-0.455	-0.646	-1.734	22.958
23	57	REF	0.587	67.822	21.384	7.896	2.311
		γ	0.568	67.290	21.530	7.720	2.900
		% DEV	-3.237	-0.784	0.683	-2.229	25.487

No.	Sample ID		%238	%239	%240	%241	%242
24	58	REF	0.783	67.632	21.383	7.896	2.306
		γ	0.562	67.510	21.330	7.780	2.830
		% DEV	-28.225	-0.180	-0.248	-1.469	22.723
25	59	REF	0.558	68.327	21.436	7.544	2.135
		γ	0.477	67.960	21.370	7.380	2.810
		% DEV	-14.516	-0.537	-0.308	-2.174	31.616
26	60	REF	0.359	69.716	21.258	6.867	1.800
		γ	0.333	69.010	21.230	6.730	2.710
		% DEV	-7.242	-1.013	-0.132	-1.995	50.556
27	62	REF	0.377	69.985	21.249	6.682	1.707
		γ	0.292	69.420	21.110	6.540	2.650
		% DEV	-22.546	-0.807	-0.654	-2.125	55.243
28	108	REF	1.077	64.270	23.190	8.355	3.108
		γ	1.029	63.920	23.020	8.380	3.650
		% DEV	-4.457	-0.545	-0.733	0.299	17.439
29	109	REF	1.081	64.321	23.142	8.350	3.106
		γ	1.042	63.820	23.190	8.230	3.720
		% DEV	-3.608	-0.779	0.207	-1.437	19.768
30	110	REF	1.115	63.441	23.725	8.429	3.290
		γ	1.089	62.900	23.620	8.420	3.960
		% DEV	-2.332	-0.853	-0.443	-0.107	20.365
31	112	REF	0.962	65.222	22.889	8.034	2.893
		γ	0.930	64.400	23.060	7.980	3.630
		% DEV	-3.326	-1.260	0.747	-0.672	25.475

No.	Sample ID		%238	%239	%240	%241	%242
32	114	REF	0.827	66.162	22.670	7.696	2.645
		γ	0.794	65.510	22.630	7.660	3.390
		% DEV	-3.990	-0.985	-0.176	-0.468	28.166
33	118	REF	0.723	66.957	22.433	7.417	2.470
		γ	0.695	66.380	22.340	7.350	3.230
		% DEV	-3.873	-0.862	-0.415	-0.903	30.769
34	119	REF	0.757	66.441	22.695	7.544	2.563
		γ	0.734	65.560	22.810	7.440	3.450
		% DEV	-3.038	-1.326	0.507	-1.379	34.608
AV DEV (%)			-4.7636	-0.3857	-0.1050	-1.4238	23.1190
ST DEV (%)			15.5562	0.4376	0.9719	0.8112	13.8776

TABLE E-3

ISOTOPIC WEIGHT RATIOS FOR PWR SAMPLES: PASSIVE γ RESULTS COMPUTED WITH MODIFIED PARAMETERS

No.	Sample ID		%238	%239	%240	%241	%242
1	4	REF	0.836	65.121	22.023	8.806	3.215
		γ	0.822	65.255	21.791	8.815	3.317
		% DEV	-1.654	0.205	-1.052	0.101	3.175
2	5	REF	1.109	62.975	22.552	9.658	3.706
		γ	1.098	62.823	22.541	9.669	3.869
		% DEV	-0.981	-0.242	-0.049	0.113	4.405
3	8	REF	0.990	63.286	22.679	9.384	3.661
		γ	0.989	63.218	22.533	9.433	3.826
		% DEV	-0.060	-0.107	-0.643	0.522	4.517
4	9	REF	0.982	62.490	23.135	9.568	3.825
		γ	0.969	62.493	22.934	9.517	4.086
		% DEV	-1.305	0.005	-0.867	-0.530	6.831
5	10	REF	1.009	62.383	23.159	9.593	3.856
		γ	0.980	62.077	23.117	9.608	4.218
		% DEV	-2.884	-0.490	-0.181	0.153	9.389
6	11	REF	0.995	62.270	23.192	9.670	3.873
		γ	0.971	61.983	23.175	9.615	4.256
		% DEV	-2.427	-0.461	-0.074	-0.568	9.898
7	12	REF	0.979	62.269	23.269	9.612	3.871
		γ	0.974	61.625	23.406	9.585	4.410
		% DEV	-0.556	-1.034	0.590	-0.282	13.921

No.	Sample ID		%238	%239	%240	%241	%242
8	13	REF	1.019	62.208	23.173	9.728	3.872
		γ	1.014	62.282	22.871	9.755	4.078
		% DEV	-0.499	0.119	-1.304	0.274	5.330
9	14	REF	1.169	61.636	23.166	10.050	3.978
		γ	1.160	61.635	23.007	9.985	4.213
		% DEV	-0.730	-0.001	-0.688	-0.649	5.904
10	22	REF	1.097	66.963	20.403	8.219	3.318
		γ	1.051	67.384	20.538	8.363	2.664
		% DEV	-4.201	0.629	0.660	1.755	-19.721
11	73	REF	1.019	63.840	21.960	9.834	3.347
		γ	0.976	63.344	22.181	9.843	3.656
		% DEV	-4.252	-0.777	1.006	0.094	9.244
12	74	REF	1.247	61.302	22.431	11.033	3.987
		γ	1.234	61.176	22.547	11.002	4.042
		% DEV	-1.067	-0.206	0.518	-0.285	1.378
13	75	REF	1.416	60.526	22.528	11.376	4.154
		γ	1.313	60.205	22.787	11.431	4.264
		% DEV	-7.270	-0.531	1.149	0.485	2.648
14	76	REF	1.331	60.638	22.524	11.604	4.102
		γ	1.291	60.644	22.447	11.567	4.051
		% DEV	-3.040	0.011	-0.342	-0.317	-1.247
15	79	REF	1.339	60.493	22.565	11.452	4.150
		γ	1.317	60.260	22.729	11.464	4.229
		% DEV	-1.631	-0.385	0.728	0.105	1.916

No.	Sample ID		%238	%239	%240	%241	%242
16	80	REF	1.324	60.619	22.529	11.401	4.127
		γ	1.305	60.351	22.620	11.557	4.166
		% DEV	-1.420	-0.443	0.406	1.372	0.952
17	81	REF	1.374	60.523	22.542	11.416	4.145
		γ	1.309	60.442	22.691	11.368	4.190
		% REF	-4.714	-0.133	0.659	-0.424	1.094
18	82	REF	1.382	60.614	22.560	11.319	4.125
		γ	1.275	60.641	22.700	11.210	4.173
		% DEV	-7.721	0.045	0.622	-0.966	1.168
19	86	REF	0.628	74.051	17.132	6.575	1.614
		γ	0.616	74.256	17.201	6.642	1.285
		% DEV	-1.950	0.277	0.402	1.026	-20.386
20	87	REF	0.564	73.228	18.146	6.405	1.657
		γ	0.525	73.198	18.272	6.396	1.609
		% DEV	-6.991	-0.041	0.697	-0.141	-2.878
21	88	REF	0.584	70.617	19.883	6.836	2.081
		γ	0.566	70.336	20.006	6.806	2.286
		% DEV	-3.039	-0.398	0.618	-0.437	9.831
22	89	REF	0.593	69.901	20.328	6.968	2.210
		γ	0.577	69.604	20.409	6.941	2.468
		% DEV	-2.673	-0.425	0.400	-0.382	11.675
23	90	REF	0.595	69.999	20.257	6.957	2.192
		γ	0.575	69.979	20.138	6.956	2.352
		% DEV	-3.325	-0.028	-0.588	-0.011	7.282

No.	Sample ID		%238	%239	%240	%241	%242
24	91	REF	0.594	69.990	20.295	6.935	2.186
		γ	0.574	69.799	20.285	6.929	2.413
		% DEV	-3.337	-0.273	-0.049	-0.087	10.376
25	92	REF	0.555	70.846	19.860	6.714	2.025
		γ	0.539	70.472	19.989	6.728	2.272
		% DEV	-2.836	-0.528	0.648	0.213	12.182
26	93	REF	0.577	70.226	20.169	6.883	2.145
		γ	0.569	69.796	20.312	6.901	2.422
		% DEV	-1.357	-0.612	0.707	0.265	12.918
27	94	REF	0.642	70.339	20.056	6.837	2.126
		γ	0.558	70.205	20.066	6.857	2.314
		% DEV	-13.048	-0.191	0.050	0.296	8.822
28	96	REF	0.598	70.152	20.182	6.909	2.159
		γ	0.570	70.027	20.125	6.933	2.344
		% DEV	-4.647	-0.178	-0.283	0.353	8.582
29	97	REF	0.551	70.944	19.842	6.661	2.002
		γ	0.526	70.738	19.811	6.727	2.198
		% DEV	-4.484	-0.290	-0.156	0.989	9.776
30	99	REF	0.537	71.431	19.551	6.569	1.912
		γ	0.514	71.211	19.572	6.609	2.094
		% DEV	-4.220	-0.308	0.108	0.605	9.524
AV DEV (%)			-3.2773	-0.2264	0.1231	0.1215	4.6168
ST DEV (%)			2.7219	0.3318	0.6332	0.6062	8.0145

TABLE E-4

ISOTOPIC WEIGHT RATIOS FOR BWR SAMPLES: PASSIVE γ RESULTS COMPUTED WITH MODIFIED PARAMETERS

No.	Sample ID		%238	%239	%240	%241	%242
1	1	REF	0.347	74.555	18.368	5.445	1.285
		γ	0.333	74.603	18.255	5.430	1.378
		% DEV	-4.035	0.065	-0.613	-0.270	7.223
2	2	REF	0.517	71.161	19.695	6.668	1.960
		γ	0.497	71.523	19.536	6.683	1.761
		% DEV	-3.902	0.509	-0.805	0.226	-10.176
3	3	REF	0.734	66.953	21.299	8.204	2.810
		γ	0.720	67.446	21.172	8.305	2.358
		% DEV	-1.964	0.736	-0.595	1.227	-16.102
4	22	REF	1.097	66.963	20.403	8.219	3.318
		γ	1.056	67.706	20.636	8.403	2.199
		% DEV	-3.743	1.110	1.141	2.241	-33.736
5	23	REF	0.720	70.349	19.638	6.956	2.337
		γ	0.701	70.752	19.720	6.994	1.834
		% DEV	-2.681	0.573	0.415	0.546	-21.534
6	25	REF	0.444	73.656	18.657	5.747	1.496
		γ	0.415	73.684	18.668	5.741	1.492
		% DEV	-6.460	0.038	0.060	-0.112	-0.278
7	26	REF	0.445	74.105	18.385	5.799	1.266
		γ	0.420	74.035	18.346	5.785	1.414
		% DEV	-5.556	-0.094	-0.211	-0.246	11.673

No.	Sample ID		%238	%239	%240	%241	%242
8	28	REF	0.451	74.764	17.841	5.836	1.108
		γ	0.793	74.553	17.579	5.829	1.246
		% DEV	75.801	-0.282	-1.471	-0.119	12.486
9	29	REF	0.465	74.640	17.640	6.134	1.121
		γ	0.416	74.731	17.507	6.118	1.228
		% DEV	-10.535	0.123	-0.754	-0.262	9.516
10	41	REF	0.544	74.816	17.409	6.074	1.157
		γ	0.423	75.007	17.274	6.120	1.176
		% DEV	-22.254	0.255	-0.776	0.756	1.673
11	43	REF	0.397	75.265	17.303	5.979	1.056
		γ	0.403	75.223	17.215	5.999	1.160
		% DEV	1.493	-0.056	-0.506	0.336	9.825
12	44	REF	0.575	72.696	18.526	6.703	1.500
		γ	0.550	72.779	18.525	6.654	1.491
		% DEV	-4.408	0.114	-0.004	-0.724	-0.568
13	45	REF	0.584	70.369	18.882	7.289	1.876
		γ	0.543	70.611	19.719	7.288	1.839
		% DEV	-7.007	0.343	4.434	-0.018	-1.954
14	46	REF	0.620	68.304	21.044	7.808	2.224
		γ	0.572	68.421	20.946	7.815	2.247
		% DEV	-7.794	0.171	-0.466	0.086	1.040
15	48	REF	0.627	67.150	21.718	8.086	2.419
		γ	0.596	67.101	21.724	8.046	2.532
		% DEV	-4.896	-0.072	0.029	-0.497	4.672

No.	Sample ID		%238	%239	%240	%241	%242
16	49	REF	0.621	67.324	21.648	8.027	2.380
		γ	0.586	67.601	21.421	7.974	2.419
		% DEV	-5.699	0.411	-1.049	-0.665	1.635
17	50	REF	0.606	67.618	21.488	7.953	2.335
		γ	0.566	67.873	21.324	7.860	2.378
		% DEV	-6.624	0.377	-0.764	-1.171	1.829
18	51	REF	0.626	67.653	21.443	7.946	2.332
		γ	0.571	67.805	21.355	7.880	2.390
		% DEV	-8.801	0.225	-0.412	-0.836	2.477
19	53	REF	0.649	67.006	21.797	8.107	2.441
		γ	0.599	67.124	21.682	8.076	2.519
		% DEV	-7.665	0.176	-0.528	-0.382	3.177
20	54	REF	0.662	66.692	21.951	8.200	2.495
		γ	0.616	66.782	21.875	8.133	2.594
		% DEV	-6.883	0.135	-0.345	-0.818	3.951
21	55	REF	0.650	66.591	22.008	8.243	2.508
		γ	0.606	66.890	21.795	8.145	2.564
		% DEV	-6.714	0.450	-0.970	-1.193	2.231
22	56	REF	0.625	66.965	21.831	8.131	2.448
		γ	0.605	67.067	21.716	8.080	2.532
		% DEV	-3.252	0.153	-0.526	-0.622	3.411
23	57	REF	0.587	67.822	21.384	7.896	2.311
		γ	0.571	67.638	21.536	7.806	2.449
		% DEV	-2.736	-0.271	0.710	-1.144	5.986

No.	Sample ID		%238	%239	%240	%241	%242
24	58	REF	0.783	67.632	21.383	7.896	2.306
		γ	0.565	67.851	21.333	7.870	2.381
		% DEV	-27.862	0.324	-0.233	-0.336	3.272
25	59	REF	0.558	68.327	21.436	7.544	2.135
		γ	0.479	68.309	21.375	7.467	2.370
		% DEV	-14.077	-0.026	-0.285	-1.021	10.988
26	60	REF	0.359	69.716	21.258	6.867	1.800
		γ	0.335	69.345	21.229	6.814	2.278
		% DEV	-6.792	-0.533	-0.137	-0.779	26.568
27	62	REF	0.377	69.985	21.249	6.682	1.707
		γ	0.293	69.750	21.107	6.624	2.226
		% DEV	-22.178	-0.336	-0.669	-0.874	30.416
28	108	REF	1.077	64.270	23.190	8.355	3.108
		γ	1.036	64.349	23.061	8.441	3.112
		% DEV	-3.816	0.123	-0.554	1.034	0.138
29	109	REF	1.081	64.321	23.142	8.350	3.106
		γ	1.049	64.254	23.234	8.288	3.176
		% DEV	-2.953	-0.105	0.397	-0.748	2.247
30	110	REF	1.115	63.441	23.725	8.429	3.290
		γ	1.097	63.364	23.678	8.470	3.391
		% DEV	-1.612	-0.122	-0.197	0.490	3.057
31	112	REF	0.962	65.222	22.889	8.034	2.893
		γ	0.936	64.830	23.101	8.040	3.093
		% DEV	-2.681	-0.601	0.925	0.080	6.904
32	114	REF	0.827	66.162	22.670	7.696	2.645
		γ	0.799	65.927	22.663	7.728	2.882
		% DEV	-3.379	-0.355	-0.030	0.421	8.969

No.	Sample ID		%238	%239	%240	%241	%242
33	118	REF	0.723	66.957	22.433	7.417	2.470
		Y	0.699	66.777	22.364	7.421	2.739
		% DEV	-3.299	-0.270	-0.308	0.058	10.897
34	119	REF	0.757	66.441	22.695	7.544	2.563
		Y	0.739	65.978	22.844	7.504	2.936
		% DEV	-2.420	-0.697	0.655	-0.530	14.537
AV DEV (%)			-4.3348	0.0762	-0.1307	-0.1725	3.4250
ST DEV (%)			15.4671	0.3816	0.9886	0.7561	11.6157

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001-025	A02	151-175	A08	301-325	A14	451-475	A20
026-050	A03	176-200	A09	326-350	A15	476-500	A21
051-075	A04	201-225	A10	351-375	A16	501-525	A22
076-100	A05	226-250	A11	376-400	A17	526-550	A23
101-125	A06	251-275	A12	401-425	A18	551-575	A24
126-150	A07	276-300	A13	426-450	A19	576-600	A25
						601-up*	A99

*Contact NTIS for a price quote.

Los Alamos