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Evaluation of TASTEX Task H: Measurement of Plutonium Isotopic Abundances by Gamma-ray Spectrometry

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Evaluation of TASTEX Task H: Measurement of Plutonium Isotopic Abundances by Gamma-ray Spectrometry

ABSTRACT

This report describes a computer-based gamma spectrometer system that we have developed for measuring isotopic and total plutonium concentrations in nitric acid solutions. The system was assembled at Lawrence Livermore National Laboratory and installed at the Tokai reprocessing plant of the Power Reactor and Nuclear Fuel Development Corporation of Japan, where it is undergoing testing and evaluation as part of the Tokai Advanced Safeguards Exercise (TASTEX). We discuss the objectives of TASTEX Task H, "High-Resolution Gamma Spectrometer for Plutonium Isotopic Analysis," and we describe the methods and equipment used, the installation and calibration of the system, and the measurements obtained from several reprocessing campaigns. In general, we find that measurements for gamma spectrometry agree well with those of mass spectrometry and of other chemical analysis. The system measures both freshly processed plutonium from the product accountability tank and aged plutonium solutions from storage tanks.

INTRODUCTION

Plutonium, as a special nuclear material and an important product of nuclear fuel reprocessing facilities, must be accurately assayed and accounted for at all times. In recognition of this fact, the governments of the United States and Japan share a common commitment to ensure the implementation of effective safeguards for plutonium. To this end, and in accordance with agreements made during September, 1977, and March, 1978, these two nations and France initiated a program to improve safeguards techniques as applied to spent-fuel reprocessing plants, with emphasis on the ap-

plicability of these techniques to the Tokai facility of the Power Reactor and Nuclear Fuel Development Corporation of Japan (PNC). Thirteen proposals were incorporated into a program plan as individual tasks known as TASTEX (the Tokai Advanced Safeguards Technology EXercise). In the statement of cooperation, the participating parties agreed to keep the International Atomic Energy Agency (IAEA) fully informed of the progress of the research and to invite agency participation in the implementation of the tasks.

PURPOSE OF TASK H

Traditional methods of plutonium analysis, e.g., mass spectrometry and gravimetric analysis, employ laboratory techniques that are performed by highly skilled chemists and technicians who work in separate analytical facilities of the plant. Such analyses do not produce immediate results that are easily monitored by an inspector. Time delays and difficulties in handling and measurement also preclude movement of samples to an alternate site for independent analysis. TASTEX Task H, entitled "High-Resolution Gamma Spectrometer for

Plutonium Isotopic Analysis," demonstrates the usefulness and accuracy of high-resolution gamma spectrometry as an alternative to chemical methods for measuring isotopic and total concentrations of plutonium in both the product loadout and storage tanks. Although Task H calls for analyses to be performed on off-line samples, the associated equipment and measurement procedures simulate many conditions that might prevail if the system were installed as an automated, in-line analysis system.

GAMMA-RAY SPECTROMETRY FOR ISOTOPIC ANALYSIS

Isotopes of plutonium are radioactive, emitting radiations of specific energy that uniquely indicate the presence of each isotope. Both alpha particles and gamma particles are emitted, with the alpha particles being the principal radiation emitted. Alpha particles are very nonpenetrating, and their measurement requires that very thin sources be prepared by means of electrodeposition or sputtering. But gamma rays are much more penetrating, and their detected pulse-height may be analyzed by a computer-based system coupled to high-resolution germanium detectors. Since these radiations are of nuclear origin, their intrinsic character is not affected by the chemical or physical form of the material. Therefore, the analysis technique may be passive and nondestructive to the sample, relying only on the detection of the gamma rays that are spontaneously emitted.

A detailed examination of plutonium gamma-ray spectra reveals that the most intense gamma rays occur at the lower energies, i.e., below 210 keV.¹ For this reason, our analysis methods make principal use of gamma rays emitted in the energy region 43-208 keV.²⁻⁵ However, the gamma-ray spectrum of a freshly separated plutonium sample, as illustrated in Fig. 1, is significantly different from a spectrum of aged material. This is because ²³⁷U and ²⁴¹Am are almost completely removed from the plutonium solution by chemical separations performed in the reprocessing plant. The most significant effect on the spectral features is the removal of the intense 59-keV peak caused by the decay of ²⁴¹Am. With ²⁴¹Am removed, the low-energy peaks of ²³⁸Pu, ²³⁹Pu, ²⁴⁰Pu, at 43, 51, and 45 keV, respectively, are very prominent. The measurement and analysis of these three peaks and of the 148-keV peak of ²⁴¹Pu form the basis for the analysis of freshly reprocessed solutions. It is also necessary to account for several small interferences in the vicinity of these peaks that are caused by radiations from ²³⁹Np, ²³⁷U, ²⁴¹Pu.

As the product solution ages, the 59-keV peak of ²⁴¹Am becomes very intense and obscures the 43-, 45-, and 51-keV peaks. The next most intense peaks are those of ²³⁸Pu, ²³⁹Pu, ²⁴⁰Pu, and ²⁴¹Pu, at 99, 129, 104.2, and 103.7 keV, respectively. However, the 99- and 104-keV peaks are unresolved components of several overlapping gamma and x-ray peaks with an energy range of 94-104 keV.

Weaker peaks for ²³⁸Pu, ²⁴⁰Pu at 152 and 160 keV have been considered, but they are not conducive to accurate analysis because of their low intensity and because of spectral interferences at the 160-keV region. Consequently the best measurement and analysis of aged solutions may be obtained by including a computational deconvolution of the many overlapping peaks in the 94-104 keV region.

The ²⁴²Pu isotope in the plutonium solutions cannot be detected by gamma-ray spectrometry. However, its abundance is usually less than 5% in first-cycle fuel, and therefore a highly accurate measure of its abundance is not required.

We have found that a good correlation exists between the abundance of ²⁴²Pu and the abundances of ²³⁹Pu, ²⁴⁰Pu, and ²⁴¹Pu. This correlation is given by the relationship

$$[242] = 52 \frac{[240][241]}{[239]^2},$$

where the bracketed quantities represent the isotopic abundances of the indicated plutonium isotopes. A study of this correlation was made at Lawrence Livermore National Laboratory using mass spectrometry data for plutonium produced in many different reactors of PWR, BWR, and CANDU design. It showed that the ²⁴²Pu abundance can generally be calculated to within $\pm 10\%$ of its measured value,⁶ as shown in Fig. 2. The above relation was therefore incorporated into the analysis programs, and the operator may select an option that makes use of it to determine ²⁴²Pu content of the sample.

DETECTOR AND SAMPLE CELL ASSEMBLY

The analysis system consists of a sample-cell assembly coupled to an intrinsic germanium detector, a number of electronic modules that process the pulses coming from the detector, and a computer system with associated interfaces and peripherals. The system acquires, stores, and interprets the spectral data. The computer-based spectrometer, without germanium detector, is shown in Fig. 3. Block diagrams of the system components of the electronics are shown in Figs. 4 and 5.

FIG. 1. Low-energy spectra of recently separated and aged plutonium solutions.

FIG. 2. Correlation of ^{242}Pu abundance with a function involving ^{239}Pu , ^{240}Pu , and ^{241}Pu . Data are from twelve reactors of PWR, BWR, and CANDU design.

FIG. 3. TASTEX computer-based gamma spectrometer.

In addition to the physical components indicated by these figures, the system contains a set of programs that control the operation of the computer. This software can be divided into three parts:

- The OS-8 package of system routines that is supplied and licensed by Digital Equipment Corporation for use on the PDP-8. These routines control the operation of the computer and the peripherals such as the terminal, disk, and line printer.

- An assembly-language program for the acquisition, display, and disk storage of spectral data.

- A set of FORTRAN programs for interpretation of the stored files of spectral data.

An operations manual⁷ provides a functional description, physical specifications, and maintenance procedures for the hardware components. Since the system consists of many hardware and software components, the operation of the spectrometer may appear to be complex. However, in

practice, most of the hardware settings are predetermined and are never changed, and others are fixed at the time of installation.

The gamma-ray detector is made of hyperpure germanium and is cryogenically cooled with liquid nitrogen. The detector is coupled to a sample-cell assembly shown in Fig. 7. The sampling arrangement, illustrated in Fig. 7, was devised so that the cell could be located outside of the glovebox and yet meet the criterion of double containment for plutonium. The cell shown in Fig. 8 consists of two containers, one inside the other. The inner cell provides the primary containment for the plutonium solution, and the outer cell prevents contamination in the event the primary containment should fail.

The body of the primary container was machined from 304 stainless steel, and the cell window consists of a 0.13-mm thick titanium inner plate that is pressure-bonded to a polycarbonate

FIG. 4. Components of the TASTEX spectrometer system.

FIG. 5. Electronics wiring diagram of the spectrometer system (ADC means analog to digital converter).

FIG. 6. Two views of the cell and detector assembly.

FIG. 7. Isotopic analysis cell and sampling system.

outer plate. Before the plates were bonded, a radioactive source of about 200 μ Ci of ^{109}Cd was electrodeposited on the surface of the titanium to serve as a reference gamma-ray source for monitoring the stability of the system. A tantalum collimator defines the sample area seen by the detector. Teflon tubes leading out from the glove box are coupled to the sample cell by means of high-pressure connectors. The secondary container was also made of 304 stainless steel; Tygon tubing surrounds the primary tubing and is connected to the glove box with specially made fittings.

A tantalum collimator defines an effective sample area of 2.5 cm^2 . Because the cell is about 0.1 cm deep, the active volume is nominally 0.25 ml; this volume ensures an adequate counting rate for solutions of the concentrations that are encountered in the plant. This configuration minimizes cell depth to reduce attenuation of the lower-energy gamma rays by the plutonium in the solution.

SPECTRUM COLLECTION AND ANALYSIS

Gamma-ray spectrometry has a wide variety of applications, but like other passive and nondestructive techniques it must be tailored to the form or type of sample being measured. The measurement conditions identified for Task H are as follows:

- Solution contains commercial-grade plutonium (50–90% ^{239}Pu).
 - Plutonium is dissolved in nitrate solutions of concentrations up to about 250 g Pu/l.
 - Measurement points are the product loadout tank (for freshly processed material), and the storage tank (for aged material).
 - Measurements are to be made off-line in the reprocessing plant analytical laboratory.
- Task H specifies highly accurate measurement ($< \pm 0.5\%$ isotopic abundances) of the $^{238-241}\text{Pu}$, and ^{241}Am when it is present, a computationally derived

FIG. 8. Components of the sample cell and secondary container.

value for ^{242}Pu , and finally the total plutonium concentration. To attain this accuracy, we first accumulate sufficient counts in a spectrum and then perform a rigorous analysis of the relevant peaks of the spectrum.

Accurate concentration measurements depend on good calibration of the system. For the TASTEX system, we require that a sample for which the isotopic abundance and concentration values are known be introduced into the counting cell. Two calibration programs, LECAL (for freshly separated samples) and PUCAL (for aged solutions) calculate the sets of constants necessary to achieve a complete calibration of the system.

Once the calibration is achieved and verified, the continued accuracy of this calibration must be assured. This is particularly important in the reduction of measurement uncertainties to less than 1%,

since detector efficiencies alone have been known to change by this amount. To monitor and correct for such efficiency changes, we have incorporated a source of radioactive ^{109}Cd into the cell window. The 88-keV gamma-ray peak from this isotope becomes an integral part of each spectrum, and the concentration calculated by means of the various plutonium peaks in the spectrum is referenced to the observed intensity of the 88-keV peak.

Because of the differences between freshly separated and aged solutions, separate analysis programs are used for the two cases. The program used for the analysis of freshly separated plutonium is called LEPA (Low-Energy Plutonium Analysis), and program PUAN (Plutonium Analysis) analyzes aged solution spectra. These programs are written in FORTRAN and they are listed in a separate report.⁸

Analysis Procedure

A 10-ml sample from the loadout or storage tank is first transferred to the glove box that is connected to the spectrometer system. The operator then draws a sample into the cell by means of the syringe assembly illustrated in Fig. 7. This first aliquot is normally used only to flush the cell and the connecting tubing and is immediately ejected into a separate container. A second sample is then drawn, and a spectrum is acquired on this sample. After sufficient data is collected, this spectrum is stored as a computer disk file. The sample is then ejected, a third sample portion is drawn, and a second spectrum taken. All analyses are duplicated in this manner to reduce source errors associated with sampling.

For freshly separated solutions, a 30-minute counting time produces results with statistical precisions of approximately 0.3% or better. Aged samples exhibit lower counting rates and require longer counting periods.

The stored data files may subsequently be recalled for reduction and interpretation by one of the two analysis programs. These programs may be in operation concurrent with the acquisition of data on a new sample. Before exiting from the analysis program, the operator may select a program option to store all of the relevant results of the analysis in an established disk file. This file is later used by the

final SUMMARY program to tabulate all the results that have been stored in it.

Spectral Peak Analysis

The first key to reduction of the complex peak groupings collected is to describe analytically the gamma-ray or x-ray peak shapes. The method and algorithms developed for calculating peak-shape profiles have been fully discussed in other reports.³⁻⁵ With this method, the peak-shape characteristics for a given detector system may be predetermined as part of the calibration process and computed for any subsequent analysis through use of a stored set of peak shape constants.

A second key to analyzing the complex multiplets produced is the generation and use of component response spectra.⁵ Such analysis is possible because both the exact gamma-ray energies and relative intensities are predetermined for each of the isotopes of interest. The peak-shape equations then allow calculation of the proper response for each of the individual gamma emissions, and these can then be linearly added to give the profile of the response of a given isotopic component to that region of the spectrum. This process greatly reduces the number of free parameters in the analysis and also produces a linear, rather than nonlinear, set of equations that can be solved by the method of least-squares analysis.

SPECTROMETER SYSTEM EVALUATION

TESTING PHASE

The spectrometer system was initially assembled and tested at Lawrence Livermore National Laboratory before installation at the Tokai reprocessing plant. Several experiments were conducted to provide expected-performance data for the system.⁹ The initial tests were designed to detect possible data bias that might be a function of changes in either the isotopic or total plutonium concentration. We studied system linearity with respect to variations of both the isotopic and total concentrations. We first analyzed a solution of aged commercial-grade plutonium diluted in a series of known increments. The results of these tests, shown in Tables 1 and 2, indicated little system bias and showed that the system could achieve the Task goal of $\pm 0.5\%$ accuracy for aged plutonium solutions.

We made a similar set of measurements using solutions from which ²⁴¹Am and ²³⁷U had been separated for the purpose of simulating freshly reprocessed plutonium solutions. For this case, we removed the cadmium absorber and used the computer program for freshly separated solutions to interpret the spectra. These results, as shown in

TABLE 1. Gamma-ray spectrometry analysis of aged plutonium solutions of different concentrations.

Concentration of solution, g Pu/l	Difference from known value, %
259	+0.17
216	-1.26
195	+0.07
131	+1.14
43	+0.20

TABLE 2. Precision of isotopic analysis by gamma-ray spectrometry of aged plutonium solutions.

Isotope	Relative abundance	Standard deviation, % ^a
²³⁸ Pu	0.236	0.32
²³⁹ Pu	76.18	0.05
²⁴⁰ Pu	19.23	0.22
²⁴¹ Pu	2.71	0.18
²⁴¹ Am	0.464	0.15

^aStandard deviation calculations are based on 27 measurements that resulted from multiple measurements at each concentration.

Tables 3 and 4, also demonstrated that the sampling and analysis systems were linear within the experimental error. In a solution of plutonium, the decay products ²⁴¹Am and ²³⁷U are continuously generated, and we therefore studied the effects of the reappearance of these isotopes as a function of time. The system showed less than 0.3% bias for measurements taken 30 days after the separation. If analyses are to be performed after 30 days, the cadmium absorber should be inserted and the sample treated as aged material.

TABLE 3. Gamma-ray spectrometry analysis of freshly processed plutonium solutions of different concentrations.

Concentration of solution, g Pu/l	Difference from known value, %
234	-0.13
195	0.05
176	+0.17
156	+0.01
117	-0.05

TABLE 4. Precision of isotopic analysis by gamma-ray spectrometry of freshly processed plutonium solutions.

Isotope	Relative abundance	Standard deviation, % ^a
²³⁸ Pu	0.236	0.27
²³⁹ Pu	76.22	0.06
²⁴⁰ Pu	19.24	0.20
²⁴¹ Pu	2.69	0.35

^aStandard deviation calculations are based on 18 measurements that resulted from multiple measurements at each concentration.

The measurements just described were all made on a single distribution of isotopic concentrations. The next test series measured solutions in which the isotopic distributions were varied systematically. These results were compared with those obtained by mass spectrometry, and the agreement was very good, as shown in Fig. 9.

During this initial testing period, the system was demonstrated to both PNC and IAEA representatives. The operating procedures were also documented in a manual for use by the spectrometer operators.⁷ The system was then disassembled and shipped to Japan for installation in the analytical laboratory of the Tokai reprocessing plant. A new counting cell was shipped because the original one became contaminated during the initial experiments.

Installation took place during September, 1979, with only minor problems being encountered. The laboratory environment: there was at first quite warm and humid, a condition that may have contributed to two minor component failures. We also found that the detector resolution was degraded by about 10 eV because of strong floor vibrations in the glove box room where it was installed.

Several installation checks were performed, including operation of diagnostic routines for the computer and the system disks, making adjustment of the pulse electronics modules, e.g., the sensitivity, gain, and zero controls, measuring the detector parameters such as resolution and peak shape, and testing the various software programs. After receiving assurance that the system was operating properly, we began the calibration program.

CALIBRATION PHASE

It was previously agreed that PNC analytical division personnel would provide the solution standards for system calibration. Since the reprocessing plant was not operating during the period November, 1978, to November 1979, it was necessary to prepare the calibration samples in the analytical laboratory. To minimize the effort made by the PNC staff members, we agreed to coordinate our calibration activities with those of Task G, since the Task H requirements for sample concentration and calibration accuracy were similar to those required by Task G.

Eighteen solution standards, prepared as three groups of six samples each, were prepared by the

FIG. 9. Deviation of measured abundance values from known values of five sample solutions of different isotopic concentration. The upper plot of each group shows the wt% of the isotope in each sample; the compositions vary left to right from weapons-grade to a typical reactor-grade distribution. The middle plot of each group shows the extent of deviation in aged samples, and the lower plot shows the deviation of the same samples after ^{241}Am and ^{237}U were removed.

workers in the analytical division. Special separations were performed on two of the sample groups to remove the ^{241}Am and ^{237}U . The third group, representing plutonium in storage, was not radiochemically purified. The eighteen samples all had the same ratio of isotopic abundances but varied in total plutonium concentration from about 50 g/l to more than 300 g/l. The concentrations of the samples were measured individually in the analytical laboratory. A minimum of two samples of each calibration solution was simultaneously measured by the gamma spectrometer system, and a total of about 75 spectra were taken during the course of the calibration exercise.

The effort required by PNC staff members to separate ^{241}Am and ^{237}U from the samples was substantial, particularly since the quantities of material required by both tasks were rather large. Because of the long time required to reconcentrate the solution

after column separation, the calibration solutions generally did not become available until about a week following separation. Therefore, the sample age did not precisely duplicate the sample condition of less than three days old that usually prevails during normal plant operation. However, this compromise was not considered serious, and some one-month-old samples were also used during the calibration exercise, although their veracity may be more questionable.

The system must be initially calibrated by means of a single standard reference solution for freshly separated and a single standard for aged solutions. After a spectrum of the reference solution is obtained, the appropriate calibration program (LECAL or PUCAL) is run, and the calibration constants are automatically determined and stored. However, we recognize that a calibration based on a single reference sample may be somewhat tenuous.

TABLE 5. Calibration-phase total plutonium concentration of freshly processed solutions as measured by gamma spectrometry and chemical analysis.

Time since separation, d	Gamma spectrometry				Chemical analysis	
	Plutonium concentration, g/l	Calculated error, %	Average concentration, g/l	Difference between duplicate analyses, %	Plutonium concentration, g/l	Difference from gamma spectrometry, %
27	49.1 48.9	(0.34) (0.21)	49.0	(0.40)	48.54	0.95
3	74.19 74.12	(0.17) (0.17)	74.15	(0.09)	73.42	0.99
21	101.7 101.4	(0.33) (0.35)	101.6	(0.30)	101.90	-0.30
3	121.1 121.2	(0.30) (0.37)	121.2	(0.16)	119.3	1.6
9	143.4 143.4	(0.26) (0.26)	143.4	(0.0)	144.0	-0.42
9	171.4	(0.35)	171.4		171.4	0.0
7	187.0 186.3	(0.28) (0.20)	186.6	(0.32)	186.6	0.0
8	196.5 197.0	(0.28) (0.21)	196.7	(0.25)	197.3	-0.31
7	216.3 216.1	(0.19) (0.26)	216.2	(0.09)	216.6	-0.18
8	220.1 220.5	(0.27) (0.27)	220.3	(0.18)	222.1	-0.36
7	253.9 253.9	(0.24) (0.27)	253.9	(0.0)	253.8	0.04
9	339.4 340.1	(0.23) (0.23)	339.7	(0.21)	338.5	0.35
			Average difference:	0.18%	Average difference:	0.46%

TABLE 6. Calibration-phase total plutonium concentration of aged solutions as measured by gamma spectrometry and chemical analysis.

Plutonium concentration, g/l	Gamma spectrometry			Chemical analysis					
	Calculated error, %	Average concentration, g/l	Difference between duplicate analyses, %	Plutonium concentration, g/l	Difference from gamma spectrometry, %				
48.3	(0.45)	48.4	(0.41)	48.12	0.60				
48.5	(0.44)								
98.1	(0.58)								
97.9	(0.48)								
97.7	(.056)	97.9	(0.41)	99.18	-1.28				
175.5	(0.57)								
202.9	(0.46)								
204.3	(0.48)								
205.2	(0.46)	204.2	(1.1)	205.0	-0.42				
204.3	(0.56)								
231.5	(0.54)								
231.7	(0.42)								
269.3	(0.46)	269.5	(0.15)	268.0	0.55				
269.7	(0.48)								
Average difference:						0.36%	Average difference:		
							0.50%		

A better and more defensible calibration may be effected by counting several or all of the standard reference solutions that have been prepared, examining the results of each based on the initial system calibration, and finally making small changes in the calibration constants to remove significant biases. The operator may easily effect these changes to the calibration files by running the DEBIAS program that is specifically supplied for this purpose.

Table 5 is a summary of the calibration results obtained for the freshly separated samples. The agreement between the gamma-ray and chemical analyses is quite good, particularly at the higher concentrations. It is not known why some of the lower concentration values do not agree more closely, but since the higher concentration range was of prime interest, the matter was not pursued. The duplicate gamma-spectrometric analyses also agree very well, with the differences also being consistent with the quoted analysis error assigned by the program.

Table 6 is a summary of the calibration results obtained for the aged solutions taken from the storage tanks. The age for this sample, as determined by the ^{241}Am ingrowth, was about 1.5 y. The overall agreement between the chemical and gamma

spectrometry results was quite acceptable, particularly in the higher concentration range.

It was also discovered that the aged solution used for calibration contained ^{237}Np and its ^{233}Pa daughter product. Small modifications were made to the analysis program to account for the presence of both of these activities.

The calibration samples all had the same ratios of isotopic abundance. It was therefore only possible to study the precision with which the various analyses reproduced this abundance ratio, and these precision values are summarized in Table 7.

TABLE 7. Precision of calibration-phase isotopic abundance measurements by gamma spectrometry.

Isotope	Relative abundance, %	Standard deviation, %	
		Freshly separated solutions	Aged solutions
Pu-238	0.430	0.28	0.40
Pr-239	73.05	0.093	0.24
Pu-240	18.369	0.30	0.63
Pu-241	6.042	0.35	0.32
Pu-242	1.658	—	—
Am-241	0.401	—	0.80

SPECTROMETER USE DURING PROCESS CAMPAIGNS

After the calibration exercise, the gamma spectrometer system was tested during several reprocessing campaigns. Here we report measurements for samples taken between December, 1979 and December, 1980, when 56 samples were taken from the product accountability tank and 38 from the storage tanks. We distinguish between these two types of samples since the first is freshly processed and the other aged—the aged samples contain ^{237}U and ^{241}Am . Analyses of the individual samples are reported in tables in the Appendix. In the following sections we summarize those tabulations and comment on the data.

ISOTOPIC ABUNDANCE MEASUREMENTS

Table 8 summarizes the isotopic abundance measurements for each of the process campaigns, combining the two types of samples although the measurement precision for aged solutions is not as good as for fresh solutions. However, comparisons with mass spectrometry are shown for each in Figs. 10 and 11.

In general, we find good agreement between the gamma spectrometry and mass spectrometry results. The most significant differences are in the

TABLE 8. Summary of isotopic abundance results.

Sample source	No. of samples		Plutonium isotope	Range of isotopic abundances, %		Average ^a bias, %	Precision σ , %
	Fresh	Aged		Low	High		
Nov.-Dec., 1979 campaign	5	2	238	0.3	1.1	-0.95	2.07
			239	63.0	74.6	0.17	0.08
			240	18.4	22.6	0.05	0.44
			241	5.4	9.7	0.15	0.21
			242	1.3	3.7	-4.54	3.59
Jan.-Feb., 1980 campaign	5	2	238	1.0	1.2	-0.96	1.05
			239	61.6	63.6	-0.007	0.16
			240	22.7	23.3	0.23	0.16
			241	9.4	10.1	-0.03	0.40
			242	3.7	4.0	-0.57	2.38
March, 1980 inventory and transfer samples	—	4	238	0.77	0.78	0.10	0.64
			239	67.0	67.1	0.18	0.21
			240	21.16	22.22	-0.22	0.38
			241	8.06	8.12	0.51	0.36
			242	2.91	2.92	-2.74	2.01
April, 1980 Demonstration samples	—	7	238		0.84	0.43	0.39
			239		65.92	-0.01	0.18
			240		21.92	0.08	0.25
			241		8.45	0.02	0.35
			242		3.16	-0.53	1.63
May, 1980 campaign	—	9	238	0.4	1.1	-7.25	4.14
			239	67.0	74.8	0.30	0.39
			240	17.6	21.7	-0.45	0.55
			241	5.1	8.2	0.14	0.89
			242	1.1	2.9	-2.32	—
June-July, 1980 campaign	18	5	238	0.4	0.9	-8.13	7.34
			239	66.6	75.3	0.21	0.22
			240	17.3	22.0	0.08	1.28
			241	6.0	8.2	-0.26	0.37
			242	1.1	2.9	-2.32	4.35

TABLE 8. (Continued.)

Sample source	No. of samples		Plutonium isotope	Range of isotopic abundances, %		Average ^a bias, %	Precision σ , %
	Fresh	Aged		Low	High		
July-Sept., 1980 Inventory and transfer samples	—	10	238	0.76	0.77	-0.93	0.56
			239	67.0	67.4	0.05	0.16
			240	21.1	21.4	0.16	0.40
			241	7.9	8.1	0.42	0.66
			242	2.76	2.84	-3.19	0.77
Sept., 1980 Campaign	8	2	238	1.0	1.3	-3.29	2.51
			239	60.4	63.6	-0.14	0.38
			240	22.3	23.0	-0.05	0.34
			241	9.7	11.6	0.79	0.69
			242	3.4	3.9	-0.16	4.17
Oct.-Nov., 1980 Campaign	12	5	238	0.5	0.6	-2.35	2.64
			239	69.8	74.2	-0.16	0.19
			240	17.1	20.4	-0.02	0.22
			241	6.4	7.2	0.70	0.51
			242	1.6	2.2	-1.04	1.84

^aIf $b = (\gamma \text{ spec} - \text{mass spec})/\text{mass spec}$, then $\bar{b} = (\sum b)/n$, and $\sigma = 100[\sum(b - \bar{b})^2/(n - 1)]^{1/2}$.

FIG. 10. Gamma spectrometry error relative to mass spectrometry for freshly separated samples.

FIG. 11. Gamma spectrometry error relative to mass spectrometry for aged samples.

^{238}Pu results. We also note that whenever a significant difference occurs, the mass spectrometry result is higher than the gamma spectrometry value. A possible explanation for the differences is the contamination with ^{238}U of the mass spectrometry source.

The results for ^{239}Pu and the ^{240}Pu compare very well. The ^{241}Pu results also agreed very well after a 2.6% bias was removed from the early data; the bias was the result of an erroneous value being used to calibrate the system. The system also measures the ^{241}Am content of the solutions. However, since this isotope is not routinely measured in the laboratory, no comparisons could be made.

The ^{242}Pu values were calculated using the correlation algorithm discussed earlier. Figure 12 shows the calculated values versus the measured values. The variation about the predicted relationship is generally less than 10% —a value

which agrees with observations made in our initial study of this relationship (see Fig. 2).⁶

Tables 9 and 10 summarize all of the isotopic results and show the average errors associated with the various analyses. Here we note that there is generally good agreement among the different indicators of the precision of measurement: the computed uncertainties (column 2), the differences between duplicate analyses (column 3), and the average deviation between gamma spectrometry and mass spectrometry (column 5). We believe this agreement gives credibility to the error values calculated by the computer program.

CONCENTRATION MEASUREMENTS

Table 11 summarizes the plutonium concentration measurements. The gamma spectrometry

FIG. 12. Difference between the calculated ^{242}Pu abundance and the abundance as measured by mass spectrometry.

TABLE 9. Differences in plutonium isotopic abundance for freshly processed solutions as measured by gamma spectrometry and mass spectrometry.

Plutonium isotope	Gamma spectrometry			
	Average calculated error, % ^a	Average difference between duplicate analyses, %	Average bias	Average difference from mass spectrometry, bias removed
238 ^b	0.23	0.16	-3.0	3.2
239	0.11	0.10	0.04	0.22
240	0.24	0.21	0.03	0.24
241	0.25	0.22	0.26	0.47
242 ^c	—	—	-3.0	3.0

^aCalculated by the LEPA analysis program based on counts in those peaks used for the analysis.

^bLarge differences in the 238 results were treated as outliers.

^cCalculated results.

values are obtained by summing the measured isotopic concentrations of masses 238, 239, 240, and 241 and adding the calculated value for ²⁴²Pu. The chemical analysis was obtained by means of a ceric oxidation method involving several steps in which the sample is initially oxidized to Pu⁴⁺ using silver oxide, reduced with a known amount of ferrous solution, finally back-titrated with ceric solution to a potentiometric endpoint.

There was fair agreement during the first six months of operation, but during later campaigns, the gamma-ray spectrometry results began to deviate from the chemical values, as shown in Table 11 and Fig. 13. It has been difficult to determine the cause of this drift. We have known for some time that the intrinsic efficiency of a germanium detector can change by one or two percent. However, we do not believe this to be the cause of

our problem because we incorporated a ¹⁰⁹Cd source into the cell window to monitor and correct for such changes in counting efficiency (see Fig. 8). Indeed, changes were observed, as shown in Fig. 14. This figure also shows data from a ¹³³Ba check source that is frequently inserted into a special slot in front of the measurement cell for additional verification of the system integrity and performance. The lower plot in Fig. 14 shows that the decay-corrected ratios of the two radioactive source activities are constant with time, confirming that the ¹⁰⁹Cd monitor source is not deteriorating or changing its gamma emission characteristics.

There is one parameter that is not monitored, and this is cell volume changes. We are currently speculating that the titanium portion of the cell window may have separated from the plastic outer portion, causing it to bow slightly inward. As a result,

TABLE 10. Differences in plutonium isotopic abundance for aged solutions as measured by gamma spectrometry and mass spectrometry.

Plutonium isotope	Gamma spectrometry			
	Average calculated error, % ^a	Average difference between duplicate analyses, %	Average bias ^b	Average difference from mass spectrometry, bias removed
238	0.7	0.5	-0.79	1.16
239	0.3	0.3	0.15	0.25
240	0.8	0.6	-0.15	0.42
241	0.6	0.2	0.40	0.63
242 ^c	—	—	-3.4	3.7

^aCalculated by the PUAN analysis program based on counts in those peaks used for the analysis.

^bLarge differences in the 238 results were treated as outliers.

^cCalculated results.

TABLE 11. Summary of plutonium concentration measurements.

Sample source	No. of samples		Pu concentration range, g/l	Average relative difference from chemical analysis, %	Standard deviation, %
	Fresh	Aged			
Nov.-Dec., 1979 campaign	5	2	212-270	-0.047	0.77
Jan.-Feb., 1980 campaign	5	2	203-245	0.86	0.73
March, 1980 inventory and transfer samples	—	7	236-254	0.24	0.72
April, 1980 Demonstration samples	—	7	155-254	0.33	1.00
May, 1980 campaign	—	9	180-220	0.11	1.01
June-July, 1980 campaign	18	5	154-248	-1.16	0.59
July-Sept., 1980 inventory and transfer samples	—	10	213-226	-2.11	1.02
Sept., 1980 campaign	8	2	133-260	-2.28	0.76
Oct.-Nov., 1980 campaign	12	5	153-235	0.50 ^a	0.89

^a A calibration adjustment of 2.3% was made before this campaign.

we would first expect the cell volume to be less than calculated from the physical dimensions, and furthermore, without any firm support of its own, the thin titanium member may be influenced to move slightly with changes in temperature or pressure, or as a result of relaxation of the teflon seal at the outer edge.

With respect to the cell volume that was expected, we did note, during the calibration exercise at Tokai, that the calibration constants which are directly related to the cell volume were approximately 25% smaller than those we had measured at LLNL when a prototype cell of the same dimensions was used. It therefore appears that the effective cell volume for the second cell is about 25% smaller than that of the prototype, even though the machined dimensions were the same. Although this observation was not understood at the time, it can perhaps now be explained by postulating that the window was bowed inward from the very beginning. Small shifts in this bowed position will also result in further changes in sample volume. Since the sample

cell is only about 0.1 cm deep, a change of 0.001 cm would cause a 1% change in the concentration measurement.

We have begun a program of monitoring the cell volume by means of sealed samples of ²⁴¹Am. A stock solution of ²⁴¹Am was prepared and placed into a number of vials and then weighed as a check of losses due to evaporation. A new vial is subsequently opened every few weeks and its contents are drawn into the analysis cell. The 59-keV peak intensity is then measured and compared with the count rate initially observed. Data from this monitoring program is not yet available.

USE OF A ¹³³Ba CHECK SOURCE

A ¹³³Ba check source was supplied as part of the spectrometer system. This source emits x rays and gamma rays at 30-, 35-, 53-, 81-, 276-, and 303-keV. It is mounted on a slide that can be inserted into a special slot in the sample cell assembly located between the detector and the cell. The ¹³³Ba

FIG. 13. Gamma spectrometry error relative to chemical analysis for three types of samples: freshly separated, aged, and those used as IAEA demonstration samples.

FIG. 14. (above) Variation over time of 81- and 88-keV peak intensities from the ^{133}Ba check-source and the ^{109}Cd monitor; (below) variation of the ratio of the 81-keV to the 88-keV peak.

provides an independent check of the system efficiency and, more importantly, of the integrity of the ^{109}Cd monitor source whose 88-keV gamma ray is also recorded in the ^{133}Ba check-source spectrum. A special program was written to analyze the six ^{133}Ba peaks and the 88-keV peak from ^{109}Cd and compare the decay-corrected values to the zero-time

value. As discussed earlier (Fig. 14), there is generally consistent agreement between the intensities of the ^{133}Ba peaks and the ^{109}Cd peak. We reiterate that the observed changes in the overall detection efficiency should not affect the plutonium concentration measurements because the internal ^{109}Cd monitor corrects the results for such changes.

CONCLUSIONS AND RECOMMENDATIONS

Our initial goal was to provide isotopic and total concentration measurements that would be accurate to within $\pm 0.5\%$. This goal was generally obtained with respect to isotopic ratio measurements, particularly for the three principal isotopes, ^{239}Pu , ^{240}Pu and ^{241}Pu . The ^{238}Pu measurements may also be very good but cannot be accurately verified by mass spectrometry because of possible mass interference by ^{238}U . The calculationally derived value for ^{242}Pu is generally determined with $\pm 10\%$ as predicted. However, some difficulty is encountered when applying the algorithm to material taken from the storage tanks. This is because the time from reactor discharge to chemical separation must be estimated to correct for the decay of ^{241}Pu ($t_{1/2} = 14.35$ y). Although this time is usually known for a given batch of fuel that is being processed, once this material is transferred to a storage tank, it becomes mixed with plutonium from other batches and loses its identity. In spite of the difficulty that results from blending various batches, the ^{242}Pu assessment is usually quite good.

As we expected, the measurements of aged samples exhibit less precision than those of freshly separated samples. This is largely a result of the greater number of counts in the peaks that are used to analyze fresh samples. Some increase in precision could be obtained by counting for a longer period of time. Also, the counting rates for aged samples are quite low since the cell volume, and hence the count rate, was optimized for the freshly separated samples.

During the course of the exercise, it became necessary to make a few minor modifications to the computer analysis program because of the observed presence of ^{237}Np and ^{239}Np in some of the samples. The PUAN program was also modified to detect a nonequilibrium condition that might exist

between ^{241}Pu and its ^{237}U daughter, particularly when freshly processed plutonium is added to the storage tank. The LEPA program was also modified to report the effective date of chemical separation based on the amount of ^{237}U and ^{241}Am ingrowth that was measured.

The results obtained for the concentration measurements of the plutonium solutions are perplexing. Although there is generally good internal consistency, particularly of replicate analyses, biases ranging up to a few percent suddenly appeared in the later results. A tentative conclusion is that the cell volume might have been changing because of creeping of the gasket material and separation of the laminated components of the cell window. We are presently trying to verify this conclusion by periodically counting a portion of a ^{241}Am monitor solution that has been prepared. If our premise is correct, a new analysis cell should be made that will include appropriate design changes to assure a more stable cell volume.

We also are concerned about the ease of maintenance of the system. At the inception of the TASTEX program, we raised the question of the type of system hardware that should be used. At that time, the desire was to install a system expeditiously, even if this meant later replacement with a more maintainable system. Such a step should now be seriously considered. One option is to replace both the Los Alamos and the LLNL computer-analyzer portion of the respective systems with a readily available commercial model that is capable of handling both detector systems. The specifications of this computer-analyzer would be such that the resulting system would be compatible with, and more maintainable by, the host laboratory. An additional advantage would be that IAEA personnel would only be required to be

familiar with one analyzer system. A disadvantage is that a hardware failure of some critical portion of the system could shut down both analysis systems unless some backup capability or redundancy of critical components was provided.

Further attention should be given to how the spectrometer system is to be implemented in IAEA safeguards procedures and what measures will be required to ensure the integrity of the system when it is left unattended in a plant. Also, procedures for calibrating the system or verifying the calibrations

are of great concern, particularly to the IAEA. Some considerations have already been given to this, but much more thought and experimental work must be invested to develop an instrument that can meet the conditions required by the IAEA.

In addition, we now need to quickly complete the transition from TASTEX experimental and testing phases to an operational capability for the IAEA. Many of the technological improvements that can be made can be implemented in parallel with operational use.

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REFERENCES

1. R. Gunnink, J. E. Evans, and A. L. Prindle, *A Re-evaluation of the Gamma Ray Energies and Absolute Branching Intensities of ^{237}C , $^{239-241}\text{Pu}$, and ^{241}Am* , Lawrence Livermore National Laboratory, Livermore, CA UCRL-52139 (1976).
2. R. Gunnink and J. F. Tinney, "Total Fissile Content and Isotopic Analysis of Nuclear Materials by Gamma-Ray Spectrometry," in: *Symposium on Analytical Methods in the Nuclear Fuel Cycle, Vienna, Austria, 1971*, International Atomic Energy Agency, Vienna (1972).
3. R. Gunnink, J. B. Niday, and P. D. Siemens, *A System for Plutonium Analysis of Solution*, Part 1: *Techniques for Analysis of Solution*, and Part 2: *Computer Programs for Data Reduction and Interpretation*, by R. Gunnink, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-51577 (1974).
4. R. Gunnink and J. E. Evans, *In-Line Measurement of Total and Isotopic Plutonium Concentrations by Gamma Ray Spectrometry*, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-52220 (1977).
5. R. Gunnink, "Gamma Spectrometric Methods for Measuring Plutonium," in: *Proceedings of the American Nuclear Society Topical Meeting, Williamsburg, VA, 1978*, National Bureau of Standard Special Publication No. 528, Washington, D.C., U.S. GPO (1978), p. 49.
6. R. Gunnink, "Use of Isotopic Correlation Techniques to Determine ^{242}Pu Abundance," *Journal of the Institute for Nuclear Materials Management* 9, 83 (1980).
7. R. Gunnink, A. L. Prindle, J. B. Niday, A. L. Van Lehn, and D. W. O'Brien, *Operations Manual for the TASTEX Gamma Spectrometer System*, Lawrence Livermore National Laboratory, Livermore California, M-106 (1979) (International Safeguards Project Office: ISPO-67).
8. R. Gunnink et al. *Computer Programs for Plutonium Isotopic Analysis of Solutions*, Lawrence Livermore National Laboratory, Livermore, CA UCID-19194 (1981).
9. R. Gunnink, A. L. Prindle, J. B. Niday, A. L. Van Lehn, and Y. Asakura, "TASTEX Gamma Spectrometer System for Measuring Isotopic and Total Plutonium Concentrations in Solutions," in: *Proceedings of the 20th Conference of the Institute for Nuclear Materials Management, Albuquerque, NM, 1979*, Vol. 8, Manhattan, Kansas, INMM (1979), p. 429.

APPENDIX. ISOTOPIC AND CONCENTRATION RESULTS OF INDIVIDUAL SAMPLE MEASUREMENTS

TABLE A1. Plutonium isotopic abundances for freshly processed solutions from the accountability tank, as measured by gamma spectrometry and mass spectrometry.

Sample No.	Isotope	Gamma spectrometry				Mass spectrometry		
		First sample abundance, wt%	Second sample abundance, wt%	Average measurement error (computed)	Average abundance	Relative difference of two samples, %	Abundance, wt%	Relative difference from gamma spectrometry, %
PH-1	Pu-238	0.3377	0.3364	0.3	0.3371	0.38	0.347	-3.0
	239	74.598	74.556	0.08	74.577	0.06	74.555	0.03
	240	18.315	18.335	0.25	18.325	0.11	18.368	0.23
	241	5.458	5.474	0.30	5.466	0.30	5.445	0.38
	242	1.29	1.30	—	1.29	—	1.285	1.0
PH-2	Pu-238	0.5072	0.5065	0.2	0.5068	0.14	0.517	-2.0
	239	71.286	71.247	0.07	71.266	0.05	71.161	0.15
	240	19.692	19.701	0.20	19.696	0.06	19.695	0.01
	241	6.659	6.680	0.22	6.670	0.31	6.668	0.02
	242	1.86	1.86	—	1.86	—	1.960	-5.4
PH-3	Pu-238	0.7250	0.7238	0.24	0.7244	0.17	0.734	-1.3
	239	67.028	67.095	0.10	67.061	0.10	66.953	0.16
	240	21.435	21.360	0.25	21.397	0.35	21.299	0.46
	241	8.218	8.233	0.25	8.220	0.18	8.204	0.20
	242	2.59	2.59	—	2.59	—	2.810	-8.5
PH-4	Pu-238	0.8216	0.8227	0.22	0.8222	0.13	0.836	-1.7
	239	65.208	65.273	0.10	65.240	0.10	65.121	0.18
	240	21.118	22.079	0.22	22.098	0.18	22.023	0.34
	241	8.814	8.802	0.24	8.808	0.14	8.806	0.02
	242	3.04	3.02	—	3.03	—	3.215	-6.1
PH-5	Pu-238	1.0967	1.0945	0.22	1.0956	0.20	1.109	-1.7
	239	63.082	63.153	0.11	63.117	0.11	62.975	0.23
	240	22.660	22.639	0.23	22.649	0.09	22.552	0.43
	241	9.678	9.652	0.24	9.665	0.27	9.658	0.07
	242	3.48	3.46	—	3.47	—	3.706	-6.8
PH-8	Pu-238	0.9837	0.9829	0.27	0.9833	0.08	0.990	-0.7
	239	63.371	63.405	0.14	63.437	0.05	63.286	0.24
	240	22.742	22.725	0.28	22.733	0.07	22.679	0.24
	241	9.405	9.398	0.30	9.451	0.07	9.384	0.71
	242	3.50	3.49	—	3.50	—	9.664	-4.7
PH-9	Pu-238	0.9732	0.9754	0.22	0.9743	0.23	0.982	-0.8
	239	62.600	62.548	0.11	62.574	0.02	62.490	0.13
	240	23.118	23.141	0.23	23.130	0.10	23.135	-0.02
	241	9.554	9.565	0.24	9.560	0.11	9.568	-0.09
	242	3.76	3.77	—	3.77	—	3.825	-1.6

TABLE A1. (Continued.)

Sample No.	Isotope	Gamma spectrometry				Mass spectrometry		
		First sample abundance, wt%	Second sample abundance, wt%	Average measurement error (computed)	Average abundance	Relative difference of two samples, %	Abundance, wt%	Relative difference from gamma spectrometry, %
PH-10	Pu-238	0.9807	0.9785	0.22	0.9796	0.22	1.009	-3.0
	239	62.303	62.317	0.11	62.310	0.01	62.383	-0.12
	240	23.266	23.254	0.22	23.260	0.05	23.159	0.44
	241	9.608	9.611	0.23	9.610	0.03	9.593	0.17
	242	3.84	3.84	—	3.84	—	3.856	-0.4
PH-11	Pu-238	0.9780	0.9804	0.23	0.9792	0.24	0.995	-1.61
	239	62.284	62.312	0.12	62.298	0.05	62.270	0.04
	240	23.288	23.238	0.23	23.263	0.22	23.192	0.31
	241	9.607	9.629	0.24	9.618	0.23	9.670	-0.54
	242	3.84	3.84	—	3.84	—	3.873	-0.8
PH-12	Pu-238	0.9738	0.9754	0.23	0.9746	0.16	0.979	-0.5
	239	62.129	62.242	0.12	62.185	0.18	62.269	-0.13
	240	23.387	23.315	0.23	23.351	0.31	23.269	0.35
	241	9.624	9.612	0.24	9.618	0.12	9.612	0.06
	242	3.89	3.85	—	3.87	—	3.870	0.0
PH-13	Pu-238	1.0183	1.0216	0.22	1.020	0.33	1.019	0.9
	239	62.285	62.134	0.12	62.210	0.21	62.208	-0.00
	240	23.137	23.237	0.23	23.187	0.43	23.173	0.06
	241	9.703	9.712	0.24	9.707	0.09	9.728	-0.21
	242	3.86	3.90	—	3.88	—	3.872	0.0
PH-14	Pu-238	1.1669	1.1654	0.24	1.1661	0.13	1.169	-0.2
	239	61.413	61.601	0.13	61.507	0.31	61.636	-0.21
	240	23.275	23.149	0.25	23.212	0.54	23.166	0.20
	241	10.024	10.014	0.26	10.019	0.10	10.050	-0.31
	242	4.12	4.07	—	4.10	—	3.978	3.3
PH-41	Pu-238	0.429	0.429	0.28	0.429	0.00	0.544	-21.14
	239	74.945	74.978	0.08	74.961	0.04	74.816	0.19
	240	17.489	17.454	0.28	17.472	0.20	17.409	0.36
	241	6.065	6.069	0.30	6.067	0.07	6.074	-0.12
	242	1.07	1.07	—	1.07	—	1.157	-7.52
PH-43	Pu-238	0.408	0.406	0.29	0.407	0.49	0.397	2.52
	239	75.354	75.355	0.08	75.354	0.00	75.265	0.12
	240	17.248	17.262	0.29	17.255	0.08	17.303	-0.28
	241	5.962	5.951	0.31	5.956	0.18	5.979	-0.38
	242	1.03	1.03	—	1.03	—	1.056	-2.46

TABLE A1. (Continued.)

Sample No.	Isotope	Gamma spectrometry				Mass spectrometry		
		First sample abundance, wt%	Second sample abundance, wt%	Average measurement error (computed)	Average abundance	Relative difference of two samples, %	Abundance, wt%	Relative difference from gamma spectrometry, %
PH-44	Pu-238	0.557	0.558	0.26	0.557	0.18	0.575	-3.13
	239	72.841	72.843	0.08	72.842	0.00	72.696	0.20
	240	18.579	18.553	0.27	18.566	0.14	18.526	0.22
	241	6.692	6.714	0.29	6.703	0.33	6.703	-0
	242	1.33	1.33	—	1.33	—	1.500	-11.33
PH-45	Pu-238	0.550	0.551	0.26	0.551	0.18	0.584	-5.65
	239	70.685	70.597	0.09	70.641	0.13	70.369	0.39
	240	19.866	19.884	0.27	19.875	0.09	19.882	-0.04
	241	7.259	7.311	0.28	7.285	0.71	7.289	-0.05
	242	1.64	1.66	—	1.65	—	1.876	-12.05
PH-46	Pu-238	0.580	0.579	0.25	0.579	0.17	0.620	-6.61
	239	68.206	68.403	0.09	68.304	0.29	68.304	0
	240	21.188	21.034	0.24	21.111	0.73	21.044	0.32
	241	7.819	7.809	0.26	7.814	0.13	7.808	0.08
	242	2.21	2.18	—	2.195	—	2.224	-1.30
PH-48	Pu-238	0.597	0.596	0.25	0.597	0.17	0.627	-4.78
	239	67.264	67.252	0.10	67.258	0.02	67.150	-0.16
	240	21.686	21.711	0.25	21.698	0.12	21.718	-0.09
	241	8.060	8.047	0.27	8.053	0.16	8.086	-0.41
	242	2.39	2.39	—	2.39	—	2.419	-1.20
PH-49	Pu-238	0.590	0.590	0.24	0.590	0.00	0.621	-4.99
	239	67.505	67.273	0.09	67.389	0.34	67.324	0.10
	240	21.583	21.570	0.23	21.666	0.77	21.648	0.08
	241	7.977	8.002	0.25	7.989	0.31	8.027	-0.47
	242	2.34	2.38	—	2.36	—	2.380	-0.84
PH-50	Pu-238	0.564	0.564	0.24	0.564	0.00	0.606	-6.93
	239	67.979	68.048	0.09	68.013	0.10	67.618	0.58
	240	21.352	21.282	0.24	21.317	0.33	21.488	-0.80
	241	7.852	7.865	0.25	7.859	0.16	7.953	-1.18
	242	2.25	2.24	—	2.245	—	2.335	-3.85
PH-51	Pu-238	0.574	0.574	0.25	0.574	0.00	0.626	-8.31
	239	67.894	67.896	0.10	67.895	0.00	67.653	0.36
	240	21.361	21.387	0.24	21.374	0.12	21.443	-0.32
	241	7.903	7.877	0.26	7.890	0.33	7.946	-0.70
	242	2.27	2.27	—	2.27	—	2.332	-2.66

TABLE A1. (Continued.)

Sample No.	Isotope	Gamma spectrometry				Mass spectrometry		
		First sample abundance, wt%	Second sample abundance, wt%	Average measurement error (computed)	Average abundance	Relative difference of two samples, %	Abundance, wt%	Relative difference from gamma spectrometry, %
PH-53	Pu-238	0.604	0.606	0.24	0.605	0.33	0.649	-6.78
	239	67.075	67.049	0.10	67.062	0.04	67.006	0.08
	240	21.793	21.795	0.23	21.794	0.01	21.797	-0.01
	241	8.097	8.112	0.25	8.105	0.18	8.107	-0.02
	242	2.43	2.44	—	2.435	—	2.441	-0.25
PH-54	Pu-238	0.623	0.622	0.24	0.622	0.16	0.662	0.00
	239	66.819	66.818	0.10	66.818	0.00	66.692	-0.23
	240	21.875	21.905	0.24	21.890	0.14	21.951	-0.28
	241	8.196	8.171	0.26	8.183	0.30	8.200	-0.21
	242	2.49	2.48	—	2.485	—	2.495	-0.40
PH-55	Pu-238	0.623	0.623	0.25	0.623	0.00	0.650	-4.15
	239	66.686	66.742	0.10	66.714	0.08	66.591	0.18
	240	21.968	21.954	0.25	21.961	0.06	22.008	-0.21
	241	8.210	8.183	0.27	8.197	0.33	8.243	-0.56
	242	2.51	2.51	—	2.505	—	2.508	-0.12
PH-56	Pu-238	0.607	0.607	0.25	0.607	0.00	0.625	-2.88
	239	67.055	67.068	0.10	67.062	0.02	66.965	0.14
	240	21.798	21.760	0.25	21.779	0.17	21.831	-0.24
	241	8.106	8.128	0.27	8.117	0.27	8.131	-0.17
	242	2.43	2.44	—	2.435	—	2.448	-0.53
PH-57	Pu-238	0.569	0.568	0.24	0.568	0.17	0.587	-3.24
	239	67.889	67.930	0.10	67.910	0.05	67.822	0.13
	240	21.409	21.339	0.24	21.374	0.33	21.384	-0.05
	241	7.869	7.899	0.26	7.884	0.38	7.896	-0.15
	242	2.26	2.26	—	2.26	—	2.311	-2.21
PH-58	Pu-238	0.568	0.568	0.23	0.568	0.00	0.783	-27.46
	239	67.890	67.823	0.09	67.857	0.10	67.632	0.33
	240	21.370	21.417	0.23	21.393	0.22	21.383	0.05
	241	7.902	7.911	0.25	7.906	0.11	7.896	0.13
	242	2.27	2.28	—	2.275	—	2.306	-1.34
PH-59	Pu-238	0.482	0.481	0.25	0.482	0.20	0.558	-13.62
	239	68.499	68.591	0.09	68.545	0.13	68.327	0.32
	240	21.366	21.303	0.24	21.334	0.29	21.436	-0.48
	241	7.498	7.485	0.26	7.492	0.17	7.544	-0.69
	242	2.16	2.14	—	2.15	—	2.135	0.70

TABLE A1. (Continued.)

Sample No.	Isotope	Gamma spectrometry				Mass spectrometry		
		First sample abundance, wt%	Second sample abundance, wt%	Average measurement error (computed)	Average abundance	Relative difference of two samples, %	Abundance, wt%	Relative difference from gamma spectrometry, %
PH-60	Pu-238	0.336	0.335	0.27	0.335	0.30	0.359	-6.69
	239	69.670	69.738	0.09	69.704	0.10	69.716	-0.02
	240	21.251	21.233	0.24	21.242	0.08	21.258	-0.08
	241	6.850	6.815	0.26	6.833	0.52	6.867	-0.50
	242	1.89	1.88	—	1.885	—	1.800	4.72
PH-62	Pu-238	0.295	0.296	0.32	0.296	0.34	0.377	-21.49
	239	70.195	70.093	0.10	70.144	0.15	69.985	0.23
	240	21.082	21.160	0.27	21.121	0.37	21.249	-0.60
	241	6.635	6.644	0.31	6.640	0.14	6.682	-0.63
	242	1.79	1.81	—	1.80	—	1.707	5.45
PH-73	Pu-238	0.994	0.993	0.25	0.993	0.10	1.019	-2.552
	239	64.060	64.042	0.12	64.051	0.03	63.840	0.331
	240	21.918	21.921	0.26	21.920	0.01	21.960	-0.182
	241	9.835	9.841	0.27	9.838	0.06	9.834	0.041
	242	3.19	3.20	—	3.20	—	3.347	-4.392
PH-74	Pu-238	1.248	1.249	0.24	1.249	0.08	1.247	0.160
	239	61.078	61.126	0.13	61.102	0.08	61.302	-0.326
	240	22.517	22.470	0.25	22.493	0.21	22.431	0.276
	241	11.159	11.169	0.25	11.164	0.09	11.033	1.187
	242	4.00	3.99	—	3.995	—	3.987	0.201
PH-75	Pu-238	1.313	1.311	0.25	1.312	0.15	1.416	-7.345
	239	60.498	60.463	0.14	60.481	0.06	60.526	-0.074
	240	22.508	22.527	0.26	22.517	0.08	22.528	-0.049
	241	11.484	11.494	0.26	11.489	0.09	11.376	0.993
	242	2.40	4.21	—	4.205	—	4.154	1.228
PH-76	Pu-238	1.303	1.307	0.26	1.305	0.31	1.331	-1.953
	239	60.493	60.424	0.15	60.459	0.11	60.638	-0.295
	240	22.494	22.500	0.28	22.497	0.03	22.524	-0.120
	241	11.510	11.546	0.28	11.528	0.31	11.404	1.087
	242	4.20	4.22	—	4.21	—	4.102	2.633
PH-79	Pu-238	1.322	1.321	0.26	1.321	0.08	1.339	-1.334
	239	60.138	60.106	0.15	60.122	0.05	60.493	-0.613
	240	22.650	22.623	0.28	22.637	0.12	22.565	0.319
	241	11.585	11.6295	0.27	11.6073	0.38	11.452	1.356
	242	4.30	4.32	—	4.31	—	4.150	3.855

TABLE A1. (Continued.)

Sample No.	Isotope	Gamma spectrometry				Mass spectrometry		
		First sample abundance, wt%	Second sample abundance, wt%	Average measurement error (computed)	Average abundance	Relative difference of two samples, %	Abundance, wt%	Relative difference from gamma spectrometry, %
PH-80	Pu-238	1.315	1.312	0.25	1.314	0.23	1.324	-0.755
	239	60.318	60.332	0.15	60.325	0.02	60.619	-0.485
	240	22.542	22.565	0.27	22.554	0.10	22.529	0.111
	241	11.568	11.544	0.27	11.556	0.21	11.401	1.360
	242	4.26	4.25	—	4.255	—	4.127	3.102
PH-81	Pu-238	1.317	1.317	0.23	1.317	0.00	1.374	-4.148
	239	60.226	60.219	0.14	60.222	0.01	60.523	-0.497
	240	22.611	22.586	0.26	22.598	0.11	22.542	0.248
	241	11.566	11.593	0.25	11.580	0.23	11.416	1.437
	242	4.28	4.29	—	4.285	—	4.145	3.378
PH-82	Pu-238	1.302	1.302	0.32	1.302	0.00	1.382	-5.789
	239	60.550	60.415	0.19	60.482	0.22	60.614	-0.218
	240	22.553	22.602	0.35	22.577	0.22	22.560	0.075
	241	11.426	11.468	0.34	11.447	0.37	11.319	1.131
	242	4.17	4.21	—	4.19	—	4.125	1.576
PH-86	Pu-238	0.620	0.618	—	0.619	0.32	0.628	-1.433
	239	74.163	74.254	—	74.208	0.12	74.051	0.274
	240	17.165	17.103	—	17.134	0.36	17.132	0.012
	241	6.645	6.628	—	6.636	0.26	6.575	0.928
	242	1.41	1.40	—	1.405	—	1.614	-12.949
PH-87	Pu-238	0.535	0.538	—	0.536	0.56	0.564	-4.965
	239	73.309	73.315	—	73.312	0.01	73.228	0.115
	240	18.210	18.170	—	18.190	0.22	18.146	0.242
	241	6.408	6.437	—	6.422	0.45	6.405	0.265
	242	1.54	1.54	—	1.54	—	1.657	-7.061
PH-88	Pu-238	0.576	0.575	—	0.575	0.17	0.584	-1.541
	239	70.542	70.495	—	70.519	0.07	70.617	-0.139
	240	19.955	19.955	—	19.955	0.00	19.883	0.362
	241	6.853	6.888	—	6.870	0.51	6.836	0.497
	242	2.07	2.09	—	2.08	—	2.081	-0.048
PH-89	Pu-238	0.589	0.590	—	0.589	0.17	0.593	-0.675
	239	69.811	69.808	—	69.810	0.00	69.901	-0.130
	240	20.368	20.340	—	20.354	0.14	20.328	0.128
	241	7.019	7.044	—	7.031	0.36	6.968	0.904
	242	2.21	2.22	—	2.215	—	2.210	0.226

TABLE A1. (Continued.)

Sample No.	Isotope	Gamma spectrometry				Mass spectrometry		
		First sample abundance, wt%	Second sample abundance, wt%	Average measurement error (computed)	Average abundance	Relative difference of two samples, %	Abundance, wt%	Relative difference from gamma spectrometry, %
PH-90	Pu-238	0.586	0.590	—	0.588	00.68	0.595	-1.176
	239	69.981	69.922	—	69.951	0.08	69.999	-0.069
	240	20.235	20.258	—	20.246	0.11	20.257	-0.054
	241	7.011	7.032	—	7.022	0.30	6.057	-0.934
	242	2.19	2.20	—	2.195	—	2.192	0.137
PH-91	Pu-238	0.585	0.587	—	0.586	0.34	0.594	-1.347
	239	69.891	69.835	—	69.863	0.08	69.990	0.181
	240	20.294	20.357	—	20.336	0.31	20.295	-0.153
	241	7.026	7.009	—	7.018	0.24	6.935	-1.197
	242	2.20	2.21	—	2.205	—	2.186	0.869
PH-92	Pu-238	0.546	0.544	—	0.545	0.37	0.555	-1.802
	239	70.851	70.969	—	70.910	0.17	70.846	0.090
	240	19.727	19.727	—	19.771	0.44	19.860	-0.448
	241	6.761	6.761	—	6.766	0.16	6.714	0.775
	242	2.02	2.00	—	2.01	—	2.025	-0.741
PH-93	Pu-238	0.575	0.576	—	0.575	0.17	0.577	0.347
	239	70.144	70.186	—	70.165	0.06	70.226	-0.087
	240	20.174	20.135	—	20.155	0.19	20.169	0.069
	241	6.954	6.957	—	6.956	0.04	6.883	1.061
	242	2.15	2.15	—	2.15	—	2.145	0.233
PH-94	Pu-238	0.569	0.569	—	0.569	0.00	0.642	-11.371
	239	70.433	70.194	—	70.313	0.34	70.339	-0.037
	240	20.044	20.162	—	20.103	0.59	20.056	-0.234
	241	6.862	6.934	—	6.898	1.05	6.837	0.892
	242	2.40	2.14	—	2.115	—	2.126	0.517
PH-96	Pu-238	0.580	0.580	—	0.580	0.00	0.598	-3.010
	239	70.157	70.151	—	70.154	0.01	70.152	-0.003
	240	20.130	20.159	—	20.144	0.14	20.182	-0.188
	241	6.979	6.959	—	6.969	0.29	6.909	0.868
	242	2.15	2.15	—	2.15	—	2.159	0.417
PH-97	Pu-238	0.542	0.541	—	0.541	0.18	0.551	-1.815
	239	70.894	71.004	—	70.949	0.16	70.944	-0.007
	240	19.838	19.755	—	19.796	0.42	19.842	0.232
	241	6.723	6.715	—	6.719	0.12	6.661	0.871
	242	2.00	1.99	—	1.995	—	2.002	0.350
PH-99	Pu-238	0.525	0.524	—	0.525	0.00	0.537	-2.235
	239	71.396	71.334	—	71.365	0.09	71.431	-0.092
	240	19.532	19.596	—	19.564	0.33	19.551	0.066
	241	6.630	6.620	—	6.625	0.15	6.569	0.852
	242	1.92	1.92	—	1.92	—	1.912	0.418

TABLE A2. Isotopic abundances for aged solutions from the storage tank, as measured by gamma spectrometry and mass spectrometry.

Sample No.	Isotope	Gamma spectrometry				Mass spectrometry		
		First sample abundance, wt%	Second sample abundance, wt%	Average measurement error (computed)	Average abundance	Relative difference of two samples, %	Abundance, wt%	Relative difference from gamma spectrometry, %
PH-6	Pu-238	0.426	0.428	0.6	0.427	0.5	0.413	3.39
	239	73.614	73.460	0.2	73.537	0.2	73.410	0.17
	240	18.166	18.330	0.7	18.248	0.9	18.360	-0.61
	241	6.155	6.142	0.4	6.149	0.2	6.155	-0.10
	242	1.66	1.66	—	1.66	—	1.662	—
	Am-241	0.432	0.431	0.4	0.432	0.2	—	—
PH-7	Pu-238	0.578	0.581	0.8	0.580	0.3	0.582	-3.34
	239	70.810	70.790	0.3	70.800	0.1	70.610	+0.27
	240	19.545	19.538	0.9	19.542	0.0	19.638	-0.49
	241	7.031	7.048	0.6	7.040	0.2	7.007	+0.47
	242	2.04	2.04	—	2.04	—	2.163	-6.0
	Am-241	0.235	0.235	0.6	0.235	0.0	—	—
PH-15	Pu-238	0.772	0.766	0.7	0.769	0.8	(a)	
	239	67.101	67.108	0.3	67.145	0.1		
	240	21.204	21.133	0.8	21.169	0.3		
	241	8.124	8.128	0.5	8.126	0.0		
	242	2.80	2.78	—	2.79	0.7		
	Am-241	0.1815	0.1821	0.6	0.1818	0.3		
PH-16	Pu-238	0.774	0.779	0.6	0.777	0.6	0.769	1.04
	239	67.011	66.899	0.3	66.955	0.2	66.975	-0.03
	240	21.215	21.256	0.8	21.236	1.2	21.217	0.09
	241	8.171	8.209	0.5	8.190	0.5	8.123	0.82
	242	2.83	2.86	—	2.845	1.0	2.916	-2.43
	Am-241	0.1955	0.1977	0.6	0.1966	1.1	—	—
PH-17	Pu-238	0.769	0.766	0.7	0.773	0.9	(a)	
	239	67.348	67.032	0.3	67.190	0.5		
	240	21.015	21.246	0.8	21.131	1.1		
	241	8.112	8.128	0.6	8.120	0.2		
	242	2.76	2.82	—	2.79	2.2		
	Am-241	0.2023	0.2030	0.6	0.2026	0.3		
PH-18	Pu-238	0.768	0.769	0.7	0.769	0.1	(a)	
	239	67.352	67.076	0.3	67.214	0.4		
	240	20.999	21.243	0.8	21.121	1.1		
	241	8.121	8.104	0.6	8.113	0.2		
	242	2.76	2.81	—	2.785	1.8		
	Am-241	0.2066	0.2071	0.6	0.2068	0.2		

TABLE A2. (Continued.)

Sample No.	Isotope	Gamma spectrometry				Mass spectrometry		
		First sample abundance, wt%	Second sample abundance, wt%	Average measurement error (computed)	Average abundance	Relative difference of two samples, %	Abundance, wt%	Relative difference from gamma spectrometry, %
PH-19	Pu-238	0.770	0.770	0.7	0.770	0.0	0.771	-0.13
	239	67.541	67.145	3	67.343	0.6	67.038	0.45
	240	20.894	21.189	0.9	21.042	1.4	21.203	-0.76
	241	8.073	8.097	0.6	8.085	0.3	8.078	0.09
	242	2.72	2.80	—	2.76	2.9	2.910	-0.05
	Am-241	0.2273	0.2267	0.6	0.2270	0.3	—	—
PH-20	Pu-238	0.776	0.772	0.7	0.774	0.5	0.775	-0.13
	239	67.159	67.119	0.3	67.139	0.1	67.088	0.08
	240	21.146	21.178	0.8	21.162	0.1	21.165	-0.01
	241	8.117	8.121	0.6	8.119	0.0	8.055	0.79
	242	2.80	2.81	—	2.805	0.3	2.917	-3.84
	Am-241	0.2403	0.2399	0.6	0.2401	0.2	—	—
PH-21	Pu-238	0.772	0.767	0.7	0.770	0.6	0.773	-0.39
	239	67.279	67.229	0.3	67.254	0.1	67.099	0.23
	240	21.100	21.131	0.9	21.116	0.1	21.158	-0.20
	241	8.076	8.087	0.6	8.082	0.1	8.055	0.34
	242	2.77	2.79	—	2.78	0.7	2.915	-4.63
	Am-241	0.2337	0.2333	0.6	0.2335	0.2	—	—
PH-22	Pu-238	1.063	1.065	0.90	1.064	0.19	1.097	-3.00
	239	66.700	67.404	0.43	67.052	1.05	66.963	+0.13
	240	20.690	20.120	1.19	20.405	2.80	20.403	0.01
	241	8.330	8.346	0.78	8.338	0.19	8.219	1.45
	242	3.23	3.08	—	3.15	—	3.318	-5.33
PH-23	Pu-238	0.710	0.713	0.85	0.712	0.42	0.720	-1.11
	239	70.639	70.724	0.33	70.682	0.12	70.349	-0.47
	240	19.646	19.592	1.05	19.619	0.27	19.638	-0.10
	241	6.985	6.966	0.67	6.976	0.27	6.956	0.29
	242	2.02	2.06	—	2.04	—	2.337	-13.99
PH-24	Pu-238	0.776	0.773	0.93	0.775	0.38	0.864	-10.30
	239	67.471	67.321	0.42	67.396	0.22	67.009	-0.58
	240	20.874	21.051	1.13	20.963	0.84	21.184	-1.04
	241	8.123	8.079	0.76	8.101	0.54	8.038	0.78
	242	2.76	2.78	—	2.77	—	2.905	-4.65
PH-25	Pu-238	0.422	0.419	0.70	0.421	0.71	0.444	-5.18
	239	73.714	73.951	0.21	73.868	0.23	73.656	0.29
	240	18.649	18.507	0.76	18.578	0.76	18.657	-0.42
	241	5.708	5.706	0.47	5.707	0.04	5.747	-0.70
	242	1.44	1.42	—	1.43	—	1.496	-4.41

TABLE A2. (Continued.)

Sample No.	Isotope	Gamma spectrometry				Mass spectrometry		
		First sample abundance, wt%	Second sample abundance, wt%	Average measurement error (computed)	Average abundance	Relative difference of two samples, %	Abundance, wt%	Relative difference from gamma spectrometry, %
PH-26	Pu-238	0.430	0.429	0.85	0.429	0.23	0.445	-3.73
	239	74.080	74.265	0.25	74.172	0.25	74.105	0.09
	240	18.515	18.353	0.93	18.434	0.88	18.385	0.27
	241	5.801	5.799	0.57	5.800	0.04	5.799	0.02
	242	1.17	1.15	—	1.16	—	1.266	-9.1
PH-27	Pu-238	0.772	0.770	0.94	0.771	0.26	0.864	-10.76
	239	67.453	67.431	0.42	67.442	0.03	67.009	0.65
	240	20.975	20.979	1.15	20.977	0.02	21.184	0.98
	241	8.053	8.066	0.77	8.060	0.16	8.038	0.27
	242	2.75	2.75	—	2.75	—	2.905	-5.34
PH-28	Pu-238	0.400	0.397	0.86	0.398	0.75	0.451	-11.7
	239	74.877	75.170	0.24	75.024	0.39	74.764	0.34
	240	17.856	17.617	0.93	17.736	1.35	17.841	0.59
	241	5.784	5.763	0.57	5.773	0.36	5.836	1.09
	242	1.08	1.05	—	1.07	—	1.108	3.55
PH-29	Pu-238	0.420	0.425	0.94	0.423	1.20	0.465	9.03
	239	74.670	74.514	0.27	74.592	0.21	74.640	-0.06
	240	17.641	17.761	1.05	17.701	0.68	17.640	0.35
	241	6.154	6.176	0.64	6.165	0.36	6.134	0.50
	242	1.11	1.12	—	1.12	—	1.121	0.0
PH-30	Pu-238	0.768	0.771	1.10	0.770	0.39	0.864	10.88
	239	67.254	67.705	0.49	67.480	0.66	67.009	0.70
	240	21.148	20.761	1.33	20.955	1.85	21.184	-1.09
	241	8.046	8.061	0.90	8.054	0.85	8.038	0.20
	242	2.78	2.68	—	2.74	—	2.905	-6.02
PH-40	Pu-238	0.776	0.769	1.09	0.773	0.91	0.864	10.53
	239	67.407	67.695	0.47	67.551	0.43	67.009	0.81
	240	20.993	20.796	1.33	20.895	0.95	21.184	1.36
	241	8.065	8.039	0.89	8.052	0.32	8.038	0.17
	242	2.76	2.70	—	2.73	—	2.905	-6.02
PH-42	Pu-238	0.770	0.770	0.75	0.770	0.00	0.781	1.43
	239	67.074	66.973	0.34	67.024	0.15	67.905	0.11
	240	21.297	21.352	1.92	21.324	0.26	21.169	0.73
	241	8.039	8.062	0.62	8.050	0.29	8.046	0.02
	242	2.82	2.84	—	2.83	—	2.907	2.72

TABLE A2. (Continued.)

Sample No.	Isotope	Gamma spectrometry				Mass spectrometry		
		First sample abundance, wt%	Second sample abundance, wt%	Average measurement error (computed)	Average abundance	Relative difference of two samples, %	Abundance, wt%	Relative difference from gamma spectrometry, %
PH-47	Pu-238	0.766	0.770	0.80	0.768	0.52	0.781	-1.66
	239	66.944	67.324	0.36	67.134	0.57	67.095	0.06
	240	21.379	21.032	0.96	21.205	1.65	21.169	0.17
	241	8.050	8.090	0.66	8.075	0.37	8.048	0.34
	242	2.85	2.78	—	2.815	—	2.907	-3.16
PH-52	Pu-238	0.767	0.776	0.82	0.771	1.17	0.777	1.30
	239	67.196	67.124	0.35	67.160	0.11	67.089	0.10
	240	21.196	21.214	0.98	21.205	0.08	21.215	0.17
	241	8.039	8.067	0.66	8.053	0.35	8.015	0.06
	242	2.80	2.82	—	2.81	—	2.904	3.45
PH-61	Pu-238	0.764	0.764	0.78	0.764	0.00	0.777	2.22
	239	67.467	67.434	0.35	67.450	0.05	67.089	0.53
	240	21.033	21.105	0.95	21.069	0.34	21.215	-0.47
	241	7.990	7.952	0.64	7.971	0.48	8.015	0.97
	242	2.75	2.75	—	2.75	—	2.904	5.71
PH-63	Pu-238	0.772	0.769	0.76	0.771	0.39	0.777	-0.772
	239	67.319	67.158	0.34	67.239	0.24	67.089	0.224
	240	21.066	21.184	0.94	21.125	1.56	21.215	0.424
	241	8.056	8.067	0.62	8.061	0.34	8.015	-1.574
	242	2.79	2.82	—	2.805	—	2.904	-3.409
PH-64	Pu-238	0.767	0.766	0.77	0.767	0.13	0.766	0.131
	239	67.350	67.266	0.35	67.308	0.12	67.366	-0.085
	240	21.139	21.200	0.94	21.168	0.29	21.173	0.024
	241	7.975	7.984	0.64	7.980	0.11	7.832	1.890
	242	2.77	2.79	—	2.78	—	2.863	-2.899
PH-65	Pu-238	0.769	0.766	0.74	0.768	0.39	0.766	0.261
	239	67.210	67.149	0.33	67.180	0.09	67.366	-0.76
	240	21.241	21.312	0.89	21.277	0.33	21.173	0.491
	241	7.981	7.964	0.60	7.975	0.21	7.832	1.826
	242	2.80	2.81	—	2.805	—	2.863	-2.026
PH-66	Pu-238	0.761	0.768	0.73	0.765	0.92	0.766	-0.131
	239	66.983	67.491	0.33	67.237	0.76	67.366	-0.191
	240	21.456	21.018	0.75	21.237	2.08	21.173	0.302
	241	7.959	7.976	0.60	7.967	0.21	7.832	1.724
	242	2.84	2.74	—	2.795	—	2.863	-2.375

TABLE A2. (Continued.)

Sample No.	Isotope	Gamma spectrometry				Mass spectrometry		
		First sample abundance, wt%	Second sample abundance, wt%	Average measurement error (computed)	Average abundance	Relative difference of two samples, ‰	Abundance, wt%	Relative difference from gamma spectrometry, ‰
PH-67	Pu-238	0.766	0.772	0.73	0.769	0.78	0.766	-0.392
	239	67.213	66.848	0.33	67.031	0.55	67.366	-0.497
	240	21.267	21.495	0.88	21.381	1.07	21.173	0.982
	241	7.954	8.006	0.59	7.980	0.65	7.832	1.890
	242	2.80	2.88	—	2.84	—	2.863	-0.803
PH-68	Pu-238	0.766	0.768	0.81	0.767	0.26	0.766	0.131
	239	67.376	67.374	0.36	67.375	0.00	67.366	0.013
	240	21.112	21.080	0.57	21.096	0.20	21.173	-0.364
	241	7.974	7.999	0.66	7.986	0.31	7.832	1.966
	242	2.77	2.78	—	2.775	—	2.863	-3.074
PH-69	Pu-238	0.767	0.753	0.77	0.760	1.86	0.766	-0.783
	239	67.379	67.292	0.34	67.335	0.13	67.366	-0.046
	240	21.148	21.285	0.93	21.216	9.65	21.173	0.203
	241	7.937	7.891	0.62	7.914	0.58	7.832	1.047
	242	2.77	2.78	—	2.775	—	2.863	-3.074
PH-70	Pu-238	0.764	0.761	0.75	0.762	0.39	0.766	-0.392
	239	67.178	67.285	0.34	67.232	0.16	67.366	-0.199
	240	21.322	21.254	0.91	21.288	0.32	21.173	0.543
	241	7.929	7.915	0.61	7.922	0.18	7.832	1.149
	242	2.81	2.78	—	2.795	—	2.863	-2.375
PH-71	Pu-238	0.760	0.761	0.74	0.762	0.13	0.766	0.783
	239	79.191	67.608	0.33	67.232	0.62	67.366	0.050
	240	21.376	21.029	0.91	21.202	1.65	21.173	0.137
	241	7.876	7.881	0.61	7.878	0.06	7.832	0.587
	242	2.80	2.72	—	2.76	—	2.863	-3.598
PH-72	Pu-238	0.758	0.758	0.81	0.758	0.00	0.766	1.044
	239	67.317	67.205	0.36	67.261	0.17	67.366	0.156
	240	21.243	21.323	0.97	21.283	0.37	21.173	0.520
	241	7.962	7.912	0.67	7.907	0.13	7.832	0.958
	242	2.79	2.80	—	2.79	—	2.863	2.550
PH-77	Pu-238	0.760	0.761	0.81	0.761	0.13	0.784	-2.934
	239	67.160	67.693	0.36	67.426	0.79	67.205	0.329
	240	21.350	20.906	0.97	21.132	2.12	21.207	-0.354
	241	7.910	7.924	0.66	7.915	0.18	7.893	0.279
	242	2.81	2.72	—	2.765	—	2.911	-5.015

TABLE A2. (Continued.)

Sample No.	Isotope	Gamma spectrometry				Mass spectrometry		
		First sample abundance, wt%	Second sample abundance, wt%	Average measurement error (computed)	Average abundance	Relative difference of two samples, %	Abundance, wt%	Relative difference from gamma spectrometry, %
PH-78	Pu-238	0.569	0.566	0.82	0.568	0.53	0.606	-6.271
	239	70.201	70.418	0.31	70.310	0.31	69.998	0.446
	240	20.143	19.984	0.94	20.064	0.80	20.224	-0.791
	241	7.198	7.176	0.62	7.187	0.31	7.134	0.743
	242	1.89	1.86	—	1.870	—	2.038	-8.243
PH-83	Pu-238	0.571	0.565	—	0.568	1.06	0.582	-1.02
	239	69.832	70.422	—	70.127	0.84	70.035	-0.01
	240	20.485	19.979	—	20.232	2.53	20.229	0.00
	241	7.173	7.174	—	7.173	0.01	7.098	1.05
	242	1.94	1.86	—	1.90	—	2.056	-8.20
PH-84	Pu-238	0.567	0.564	—	0.565	0.53	0.582	-1.03
	239	70.285	70.327	—	70.306	0.06	70.035	0.04
	240	20.130	20.101	—	20.115	0.14	20.229	-0.57
	241	7.144	7.140	—	7.142	0.06	7.098	0.62
	242	1.87	1.87	—	1.87	—	2.056	-10.00
PH-85	Pu-238	0.575	0.577	—	0.576	0.35	0.582	-1.01
	239	69.906	69.920	—	69.913	0.02	70.035	-0.02
	240	20.366	20.361	—	20.363	0.02	20.229	0.29
	241	7.215	7.208	—	7.212	0.10	7.098	1.53
	242	1.941	1.93	—	1.935	—	2.056	-6.80
PH-95	Pu-238	0.577	0.580	—	0.579	0.52	0.582	-1.00
	239	69.966	70.031	—	69.998	0.09	70.035	-0.01
	240	20.313	20.262	—	20.287	0.25	20.229	0.29
	241	7.210	7.204	—	7.207	0.08	7.098	1.53
	242	1.93	1.92	—	1.925	—	2.056	-6.80
PH-98	Pu-238	0.573	0.571	—	0.574	0.35	0.582	-1.37
	239	70.044	69.981	—	70.012	0.09	70.035	-0.03
	240	20.359	20.297	—	20.328	0.31	20.229	0.48
	241	7.115	7.214	—	7.164	1.39	7.098	0.93
	242	1.91	1.93	—	1.92	—	2.056	-6.61

^aMass spectrometry measurement was performed on these samples since the plant was not operating during this period. The isotopic data for PH-16 may be assumed to apply.

TABLE A3. Total plutonium concentration for freshly processed solutions as measured by gamma spectrometry and by chemical analysis with the ceric titration method.

Sample No.	Gamma spectrometry				Chemical analysis	
	First sample, g/l (calculated error)	Second sample, g/l (calculated error)	Average concentration, g/l	Relative difference, %	Average concentration, g/l	Relative difference from gamma spectrometry, %
PH-1	212.92 (0.18)	212.72 (0.23)	212.82	0.09	212.1	0.34
PH-2	254.44 (0.13)	254.36 (0.17)	254.4	0.03	255.9	-0.59
PH-3	230.51 (0.20)	230.31 (0.20)	230.41	0.09	231.2	-0.34
PH-4	269.30 (0.19)	268.81 (0.19)	269.05	0.18	269.6	-0.20
PH-5	250.56 (0.19)	250.74 (0.19)	250.64	0.08	250.5	-1.46
PH-8	205.96 (0.23)	205.84 (0.23)	205.90	0.06	203.2	1.33
PH-9	233.93 (0.19)	233.25 (0.19)	233.58	0.29	230.1	1.51
PH-10	241.35 (0.18)	241.60 (0.18)	241.47	0.10	237.9	1.50
PH-11	244.74 (0.20)	243.96 (0.20)	244.35	0.32	245.5	-0.47
PH-12	242.50 (0.19)	242.11 (0.19)	242.30	0.15	240.1	0.92
PH-13	232.69 (0.19)	232.67 (0.19)	232.68	0.01	233.3	0.26
PH-14	225.18 (0.21)	225.60 (0.21)	225.39	0.18	223.1	1.03
PH-41	183.2 (0.22)	182.8 (0.22)	183.0	0.22	185.1	-1.13
PH-43	157.5 (0.22)	157.7 (0.22)	157.6	0.13	160.1	-1.56
PH-44	182.5 (0.21)	181.9 (0.21)	182.2	0.33	183.8	-0.87
PH-45	180.6 (0.21)	179.9 (0.21)	180.3	0.39	182.1	-0.99
PH-46	201.12 (0.20)	200.7 (0.20)	200.9	0.20	202.8	-0.94
PH-48	190.0 (0.21)	189.9 (0.21)	190.0	0.05	191.8	-0.94
PH-49	215.8 (0.19)	215.5 (0.19)	215.7	0.14	217.4	-0.88
PH-50	225.4 (0.19)	225.4 (0.20)	225.4	0.00	227.6	-0.97
PH-51	214.3 (0.20)	214.1 (0.20)	214.2	0.09	215.7	-0.69
PH-53	218.5 (0.19)	218.5 (0.19)	218.5	0.00	221.7	-1.44
PH-54	208.2 (0.20)	208.1 (0.20)	208.2	0.05	210.6	-1.14
PH-55	204.2 (0.21)	204.3 (0.21)	204.3	0.05	208.1	-1.83

TABLE A3. (Continued.)

Sample No.	Gamma spectrometry				Chemical analysis	
	First sample, g/l (calculated error)	Second sample, g/l (calculated error)	Average concentration, g/l	Relative difference, %	Average concentration, g/l	Relative difference from gamma spectrometry, %
PH-56	207.3 (0.21)	207.4 (0.21)	207.4	0.05	210.3	-1.38
PH-57	213.4 (0.20)	213.3 (0.20)	213.4	0.05	219.1	-2.60
PH-58	243.0 (0.19)	242.6 (0.19)	242.8	0.16	247.3	-1.82
PH-59	226.2 (0.20)	225.9 (0.20)	226.1	0.13	226.9	-0.35
PH-60	218.5 (0.19)	218.9 (0.19)	218.7	0.18	222.5	-1.71
PH-62	152.1 (0.22)	152.3 (0.22)	152.2	0.13	153.8	-1.04
PH-73	172.9 (0.22)	173.4 (0.22)	173.1	0.29	177.1	-2.26
PH-74	226.0 (0.23)	225.7 (0.22)	225.7	0.26	230.2	-1.95
PH-75	214.1 (0.23)	214.0 (0.23)	214.0	0.05	218.9	-2.24
PH-76	187.2 (0.24)	187.1 (0.24)	187.2	0.05	192.1	-2.55
PH-79	207.8 (0.24)	207.4 (0.24)	207.6	0.19	213.5	-2.76
PH-80	215.4 (0.24)	215.9 (0.24)	215.6	0.19	220.3	-2.13
PH-81	254.8 (0.22)	255.0 (0.22)	254.9	0.08	259.8	-1.89
PH-82	127.6 (0.30)	127.8 (0.30)	127.7	0.16	133.2	-4.13
PH-86 ^a	163.6	163.9	163.8	0.18	163.5	0.18
PH-87	223.2	222.4	222.8	0.36	220.0	1.27
PH-88	159.7	159.7	159.8	0.00	159.4	0.25
PH-89	153.2	153.5	153.4	0.19	153.2	0.13
PH-90	233.8	233.5	233.6	0.13	233.5	0.04
PH-91	230.7	230.6	230.6	0.04	231.0	-0.17
PH-92	233.9	233.7	233.8	0.08	235.3	-0.64
PH-93	220.1	220.2	220.2	0.04	220.5	-0.14
PH-94	197.3	197.5	197.4	0.10	196.5	0.46
PH-96	215.9	216.0	216.0	0.05	213.5	1.17
PH-97	201.1	201.0	201.1	0.05	198.6	1.26
PH-99	225.0	225.4	225.4	0.31	219.8	2.55

^a A calibration adjustment of 2.3% was made between the time of sample PH-82 and sample PH-86.

TABLE A4. Total plutonium concentration for aged solutions as measured by gamma spectrometry and by chemical analysis using the ceric titration method.

Sample No.	Gamma spectrometry				Chemical analysis	
	First sample, g/l (calculated error)	Second sample, g/l (calculated error)	Average concentration, g/l	Relative difference, %	Average concentration, g/l	Relative difference from gamma spectrometry, %
PH-6	216.60 (0.49)	216.46 (0.41)	216.5	0.1	217.1	-0.30
PH-7	244.58 (0.63)	244.98 (0.56)	244.9	0.1	244.4	+0.24
PH-15	255.66 (0.58)	255.99 (0.61)	255.83	0.1	253.8	0.80
PH-16	238.63 (0.61)	238.71 (0.54)	238.67	0.0	240.4	-0.72
PH-17	240.72 (0.60)	241.21 (0.61)	240.97	0.2	241.0	-0.01
PH-18	241.50 (0.60)	241.52 (0.61)	241.51	0.0	243.1	-0.65
PH-19	237.88 (0.61)	237.34 (0.64)	237.61	0.2	235.8	0.77
PH-20	239.45 (0.59)	239.31 (0.61)	239.38	0.1	236.9	1.05
PH-21	243.12 (0.63)	243.16 (0.64)	243.14	0.0	242.0	0.47
PH-22	182.64	182.21	182.43	0.23	184.14	-0.94
PH-23	218.01	217.70	217.86	0.14	216.45	0.65
PH-24	217.73	218.57	218.15	0.38	221.30	-1.42
PH-25	222.09	222.21	222.15	0.05	220.35	0.81
PH-26	220.34	220.76	220.55	0.19	219.7	0.67
PH-27	224.65	222.94	224.30	0.32	224.45	-0.05
PH-28	217.83	218.16	218.16	0.30	215.58	1.11
PH-29	168.16	168.58	168.37	0.25	169.44	-0.64
PH-30	224.32	224.12	224.22	0.09	223.98	0.11
PH-40	225.6 (0.98)	226.8 (0.84)	226.2	0.53	225.6	0.27
PH-42	227.9 (0.70)	227.8 (0.61)	227.9	0.04	229.5	-0.70
PH-47	232.1 (0.75)	231.4 (0.63)	231.7	0.38	236.0	-1.82
PH-52	236.0 (0.74)	236.0 (0.65)	236.0	0.00	239.6	-1.50
PH-61	213.6 (0.65)	213.9 (0.69)	213.8	0.14	215.3	-0.697
PH-63	214.8 (0.63)	216.6 (0.71)	215.7	0.84	219.7	-1.85
PH-64	218.9 (0.68)	216.6 (0.67)	217.7	1.06	220.7	-1.35
PH-65	219.3 (0.65)	219.7 (0.65)	219.5	0.18	224.0	-2.01
PH-66	219.7 (0.62)	220.8 (0.66)	220.2	0.50	221.8	-0.72
PH-67	219.8 (0.65)	219.7 (0.62)	219.8	0.05	224.2	-1.96

TABLE A4. (Continued.)

Sample No.	Gamma spectrometry				Chemical analysis	
	First sample, g/l (calculated error)	Second sample, g/l (calculated error)	Average concentration, g/l	Relative difference, %	Average concentration, g/l	Relative difference from gamma spectrometry, %
PH-68	219.4 (0.72)	221.6 (0.65)	220.5	1.00	225.8	-2.4
PH-69	207.4 (0.69)	207.9 (0.72)	207.6	0.24	215.1	-3.49
PH-70	208.8 (0.66)	207.9 (0.65)	208.4	0.43	216.2	-4.53
PH-71	210.9 (0.70)	210.4 (0.65)	210.7	0.24	213.3	-1.23
PH-72	209.8 (0.64)	210.0 (0.65)	209.9	0.10	214.7	-2.24
PH-77	215.7 (0.69)	215.4 (0.71)	215.6	0.14	218.9	-1.51
PH-78	229.4 (0.65)	229.5 (0.67)	229.4	0.04	232.8	-1.46
PH-83 ^a	221.8	222.1	222.0	0.14	223.2	-0.538
PH-84	227.5	228.1	227.8	0.26	224.4	1.515
PH-85	225.1	225.5	225.3	0.18	225.2	0.044
PH-95	234.6	234.2	234.4	0.17	235.4	-0.425
PH-98	240.8	239.3	240.1	0.63	236.4	1.565

^a A calibration adjustment of 2.3% was made between the time of sample PH-78 and sample PH-83.