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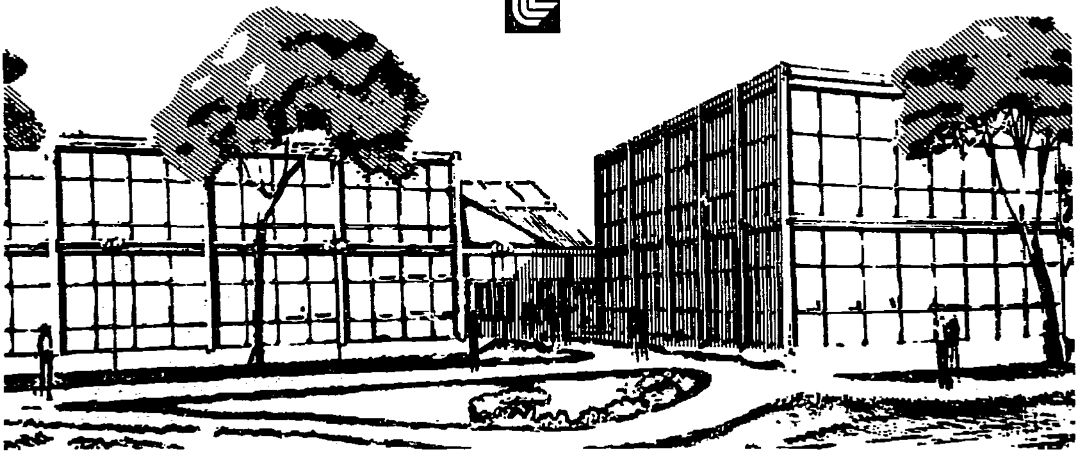

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TASTEX* Gamma Spectrometer System for Measuring Isotopic and Total Plutonium Concentrations in Solutions

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ABSTRACT

We describe a computer-based gamma ray spectrometer system using a germanium detector for rapid nondestructive measurement of isotopic and total plutonium concentrations in solutions at nuclear reprocessing plants. We have measured isotopic concentrations with an accuracy of $\pm 0.5\%$. We discuss cell design, calibration techniques, and preliminary results. This system is being installed at the Tokai reprocessing plant in Japan.

INTRODUCTION

The accountability and safeguarding of plutonium as a special nuclear material continues to be an important issue, and International Atomic Energy Agency (IAEA) inspectors need a rapid analytical method to determine isotopic and total plutonium concentrations in solutions at nuclear reprocessing plants that is not dependent on the analytical facilities at the plant. Traditional chemical methods for plutonium analysis remain indispensable, but they are sample-destructive, manpower intensive, and time-consuming. As a result, non-destructive instrumental methods are assuming an increasing role in providing rapid material balance and control of plutonium inventories. To be useful, such instrumental methods must not only determine the total amount of plutonium but the isotopic breakdown (^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, and ^{241}Am) as well, which is also frequently important.

We have developed a spectrometer system that, when properly calibrated, provides rapid and non-destructive analyses of solutions containing these isotopes. Because all of these isotopes (except ^{242}Pu) emit detectable gamma rays,[1] we developed a spectrometer system that uses a small but very-high-resolution germanium detector. Only the 40-210 keV region of the

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*TASTEX (Tokai Advanced Safeguards EXercise) is a cooperative program involving the United States, Japan, and the IAEA for testing advanced safeguards instrumentation at the Tokai reprocessing plant. The Lawrence Livermore Laboratory (LLL) portion is sponsored by the U.S. Department of Energy under contract No. W-7504-ENG-48 and administered through the International Safeguards Project Office at Brookhaven, New York.

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germanium spectrum is analyzed since the most intense and, in a few cases, the only useable gamma signals for spectroscopic analysis occur in this region. Because some of the peaks are very complex and overlap extensively, a minicomputer is required to interpret the spectra.

This instrumental method has evolved over several years, but, although many experiments have been performed and several systems are in use,[2,3] there have been few opportunities to test this method in an operating plant having sizeable inventories of reactor-grade plutonium. However, because of the commitment by the United States to strengthen IAEA capabilities and ensure effective IAEA safeguards, the International Safeguards Projects Office (ISPO) has supported our effort to develop and install a gamma spectrometer system at the Tokai-mura reprocessing plant in Japan. Using this system, IAEA inspectors will be able to verify the amounts of plutonium in storage and accountability tanks rapidly without requiring an analytical laboratory.

SAMPLE CELL DESIGN AND SAMPLING METHOD

At the Tokai reprocessing plant, the measurement points of initial interest are the product accountability and storage tanks. Routine samples obtained from these sources are sent pneumatically in special containers to the analytical laboratory where they are received in glove boxes. These nitric acid solutions contain about 200-250 g of plutonium/l.

We devised a sampling arrangement so that our sample cell could be located outside the glove box and still meet the double-containment criterion. Figure 1 shows that the cell assembly consists of two containers, one inside the other. The inner primary container holds the plutonium solution, and the outer secondary container prevents contamination of the laboratory if the primary container 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 pressure-bonded to a polycarbonate outer plate. Before the plates are bonded, a layer of radioactive ^{109}Cd is electrodeposited on the surface of the titanium bonded to the plastic 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[4] sample tubes leading from the glove box are coupled to the sample cell by high-pressure connectors.

The secondary container was also made of 304 stainless steel; Tygon[4] tubing surrounds the primary tubing and is connected to the glove box with specially made fittings.

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

During an analysis, the operator withdraws solution into the cell from a suitable container using a syringe assembly (Fig. 2). This cell design and sampling method is only one step removed from a truly in-line sampling and

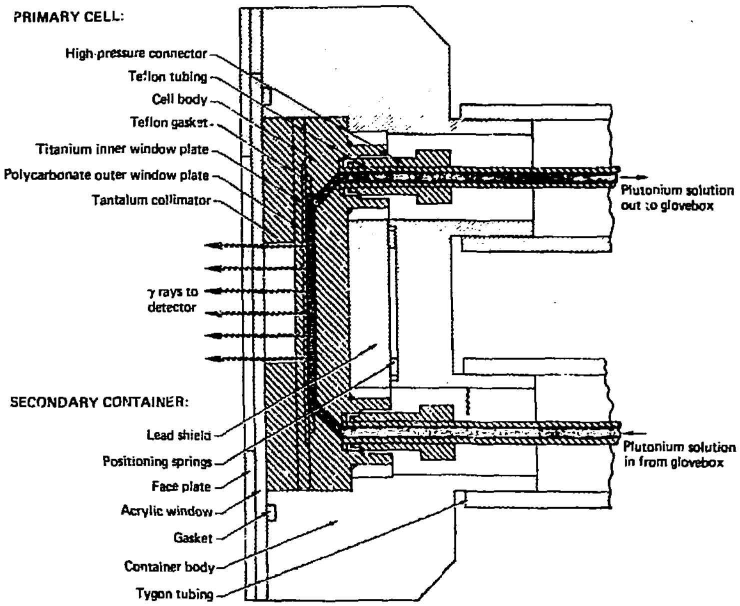

Fig. 1 Sample cell and secondary container.

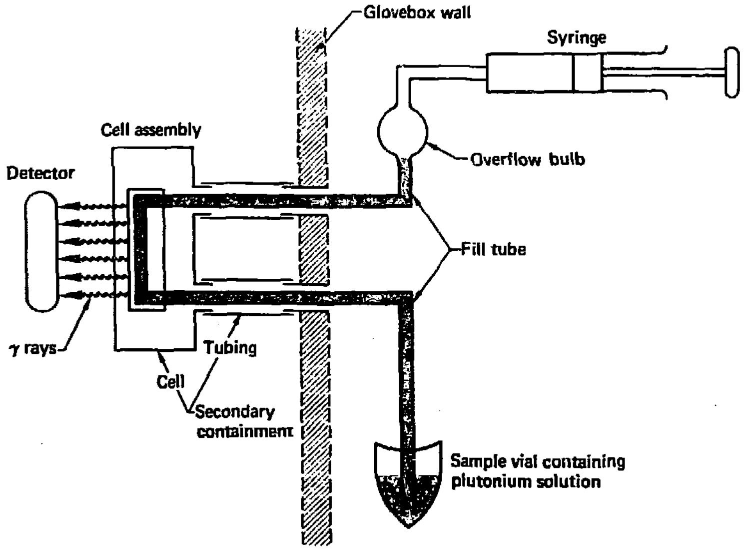

Fig. 2 Isotopic analysis cell and sampling system.

analysis procedure; in the future, this system could be incorporated directly into the reprocessing stream and the isotopic concentrations determined automatically without human intervention.

DATA ACQUISITION SYSTEM

Gamma rays from the sample cell and the ^{109}Cd monitor source are detected by a high-purity germanium detector with a nominal volume of 1 cm^3 and a 510-eV FWHM resolution at 122 keV. In addition to the usual necessary electronic modules associated with the pulse-height analyser system, we included 1) a pulse pile-up and live-time-corrector module to reduce the number of chance coincidence pulses and correct the live-time clock and 2) a spectrum stabilizer to prevent drift in the system gain or zero levels caused by electronic instabilities arising from changes of counting rate and temperature.

We use an analog-to-digital converter interfaced with a PDP-8/E computer to digitize and store incoming pulses from the germanium detector; a 4,096-channel spectrum is taken at a system gain of 0.075 keV/channel. Once data acquisition has been completed, the spectrum is stored on a magnetic disc cartridge. Spectra are subsequently analyzed by specifically designed computer programs that reduce and interpret the data and calculate plutonium isotopic concentrations.

DATA ANALYSIS

All of the isotopes of interest except ^{242}Pu emit observable gamma- and x-radiations in the low-energy region of a germanium gamma-ray spectrum. However, the spectra of recently processed plutonium solutions differ considerably from spectra of solutions that have been aged a month or more. The latter contain ^{241}Am and ^{237}U, which are daughter products of ^{241}Pu.

For plutonium separated within the previous three or four weeks, we used the 43.48 keV peak in the germanium spectrum to measure ^{238}Pu, the 51.63 keV peak for ^{239}Pu, the 45.23 keV peak for ^{240}Pu, and the 148.57 keV peak for ^{241}Pu. Because the region between 43 and 46 keV contains small interferences from ^{239}Pu and ^{241}Pu, careful analysis of the ^{238}Pu and ^{240}Pu peaks (43.48 and 45.23 keV, respectively) was required. We also analyzed the 129.28 and 94.66 keV peaks to confirm the results obtained for ^{239}Pu and ^{241}Pu, respectively.

The spectra of aged solution generally exhibit an intense ^{241}Am peak at 59.54 keV that can be reduced using a 0.1-cm Cd absorber. However, spectral features of lower energy are obscured by the Compton continuum, and it was therefore necessary to analyze the complex multiplet in the region between 94 and 104 keV for ^{238}Pu and ^{240}Pu isotopic data. This multiplet arises from x rays and gamma rays of up to 14 different energies. To analyze this region, we developed algorithms that accurately describe both x- and gamma-ray line shapes.[5] We also used the areas of peaks of other energies to analyze aged solutions: 59 keV (^{241}Am), 129 keV (^{239}Pu), 148 keV (^{241}Pu), 152 keV (^{238}Pu), and 208 keV (^{237}U).

We wrote separate computer programs to analyze recently processed and aged plutonium solutions. The former program is less complex than the latter one because the latter performs a least-squares solution of 140 equations to resolve the complex 94-104 keV region. Each program calculates concentration results in two ways: 1) on the basis of the counts recorded during the period of time that has elapsed according to the live-time clock, and 2) on the count rate of the 88-keV peak of the internal ^{109}Cd monitor. The latter method is considered more accurate because it automatically corrects for small changes in detector efficiency (about 1%) and corrects for small errors resulting from timing inaccuracies, particularly when dead times approach 40%.

We have also developed separate computer programs to calibrate the system for the two types of plutonium-containing solutions discussed above. These programs require minimal input and interaction by the operator. A spectrum of a sample of known isotopic composition (e.g., an NBS or other appropriate standard) is taken by the system, and the calibration program is run. To calibrate the system, the operator enters the isotopic composition of the known sample and certain cell parameters; the computer calculates the coefficients required to interpret unknown spectra and stores these coefficients as a disk file.

Gamma spectrometry cannot determine the ^{242}Pu content of plutonium samples in practice, but the effects of this limitation are not severe. First, ^{242}Pu concentrations seldom exceed 5% in first-cycle reactor fuel. Second, because a reprocessing plant generally requires several weeks to process a single reactor core, the isotopic composition during a campaign does not vary greatly; once the stream average value of ^{242}Pu has been determined by mass spectrometry, it can be applied to all samples coming from that core assembly material. Third, calculation of the ^{242}Pu abundance from the measured abundances of the other isotopes appears likely. We have found that an adequate correlation is given by the expression

$$\left[^{242}\mathrm{Pu}\right] = C\,\frac{\left[^{240}\mathrm{Pu}\right]\left[^{241}\mathrm{Pu}\right]}{\left[^{239}\mathrm{Pu}\right]^2} ,$$

where the bracketed quantities are the respective abundances at the time the fuel elements are removed from the reactor and C equals about 53. A cursory investigation indicated that this expression works quite well for plutonium produced in PWR and BWR reactors with standard fuel loadings, and we intend to study this methodology in greater detail.

Our analysis programs incorporate three options for dealing with ^{242}Pu abundance. They can use a ^{242}Pu abundance specified by the operator, retrieve the stream average value already on file, or calculate the abundance using the above equation.

PRELIMINARY RESULTS

Before installation of the system at the Tokai reprocessing plant, we performed calibration and experimental tests to provide performance expectation data for the system. Our initial tests were devised to discern any systematic biases resulting from changing plutonium concentration, and we studied the system linearity with respect to variations of both the isotopic and total concentrations.

In the first measurement series, we analyzed a solution of aged reactor-grade plutonium diluted by known increments. The results indicated that very little bias existed and that the system could achieve the anticipated goal of ±0.5% accuracy for aged plutonium solutions (Tables 1 and 2).

We made a similar set of measurements using solutions from which ^{241}Am and ^{237}U had been separated to simulate freshly reprocessed plutonium solutions. In this case, we removed the cadmium absorber and used the computer program for freshly separated solution to interpret the spectra. These results demonstrated that, within experimental error, potential sources of serious bias did not affect the linear response the system (Tables 3 and 4).

Because the decay products ^{241}Am and ^{237}U are continuously generated in a plutonium-containing solution, we studied the effects of the reappearance of these isotopes as a function of time. The plutonium measurements showed less than 0.3% bias for measurements taken 30 days after isotope separation. If analyses are to be performed after 30 days, the cadmium absorber should be reinserted and the sample treated as aged material.

The above measurements were all made using a single distribution of isotopic concentrations. We also measured a series of solutions in which the isotopic distributions were varied systematically. We compared the results measured by the TASTEX spectrometer system with those obtained by mass spectrometry and found very little deviation with different isotopic distributions (Figs. 3a-d).

A thorough performance evaluation of the system will be completed at the Tokai reprocessing facility. This evaluation will compare the measurement accuracy with standard analytical analyses and investigate the effects of such experimental parameters as changing isotopic percentages, total concentrations, count time, and fission product impurities. We hope to acquire experimental data and experience relevant to the automation and ultimate in-line installation of this measurement system. We also intend to study problems related to tampering with the system hardware and software.

CONCLUSIONS

The initial system checkout indicated that analyses for $^{238-241}Pu$ and ^{241}Am using gamma ray emissions can be made in less than an hour with an accuracy of ±0.5%. Measurements showed that, within experimental error, the system response was linear and unaffected by changes in plutonium concentration, relative isotopic abundances, and the presence or absence of decay products (specifically ^{241}Am and ^{237}U). The concentration of ^{242}Pu, which cannot be detected by gamma spectrometry, can be determined by alternate complementary techniques.

Table 1. Analysis of aged plutonium solutions of different concentrations.

Concentration of solution, g Pu/l	Difference,[a] %
259	+0.17
216	-0.26
195	+0.07
131	+0.14
43	+0.20

[a]Difference between measured and known values.

Table 2. Precision of isotopic analysis 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

[a]Standard deviation calculations based on 27 measurements that resulted from multiple measurements at each concentration.

Table 3. Analysis of freshly processed plutonium solutions of different concentrations.

Concentration of solution, g Pu/l	Difference,[a] %
234	-0.13
195	-0.05
176	+0.17
156	+0.01
117	-0.05

[a]Difference between measured and known values.

Table 4. Precision of isotopic analysis 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

[a]Standard deviation calculations based on 18 measurements that resulted from multiple measurements at each concentration.

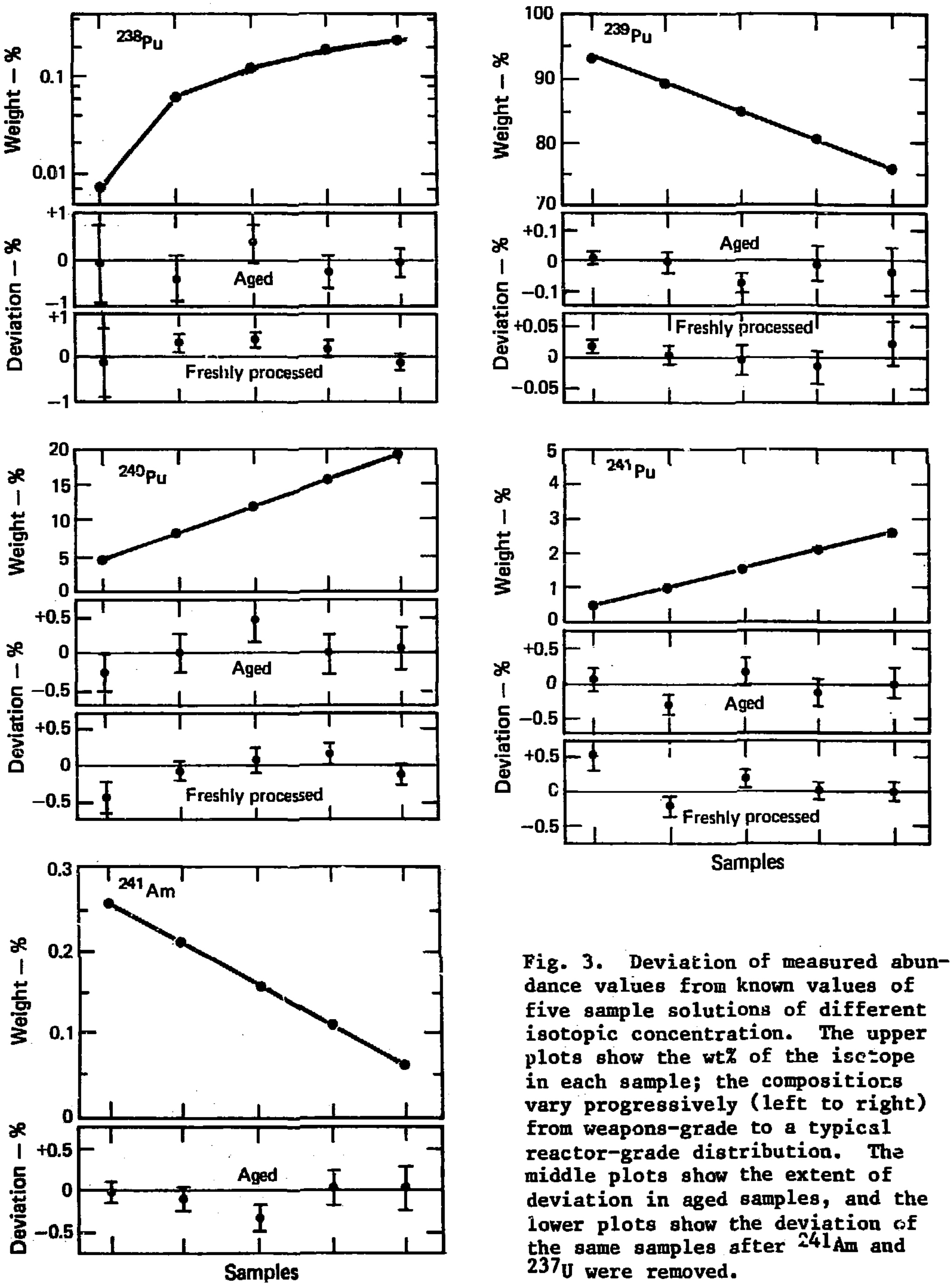

Fig. 3. Deviation of measured abundance values from known values of five sample solutions of different isotopic concentration. The upper plots show the wt% of the isotope in each sample; the compositions vary progressively (left to right) from weapons-grade to a typical reactor-grade distribution. The middle plots show the extent of deviation in aged samples, and the lower plots show the deviation of the same samples after ^{241}Am and ^{237}U were removed.

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