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A NONDESTRUCTIVE ASSAY INSTRUMENT FOR MEASUREMENT OF PLUTONIUM IN SOLUTIONS

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ABSTRACT

A nondestructive assay (NDA) instrument that measures the ^{239}Pu content in solutions, using a passive gamma-ray spectroscopy technique, has been developed and installed in the Plutonium Processing Facility at Los Alamos Scientific Laboratory (LASL). A detailed evaluation of this instrument has been performed. The results show that the instrument can routinely determine ^{239}Pu concentrations of 1 to 500 g/ℓ with accuracies of 1 to 5% and assay times of 1 to 2×10^3 s.

INTRODUCTION

Plutonium solutions are generated by a variety of chemical processes in the Los Alamos Scientific Laboratory (LASL) Plutonium Processing Facility. These solutions include: products of impure oxide metal scrap dissolutions, ion-exchange column eluates and effluents, and precipitation filtrates. The plutonium concentrations vary over a wide range, from a few mg/ℓ to over 500 g/ℓ. The solutions also contain fission products and daughter isotopes, in particular, ^{241}Am and ^{237}U. The density of ^{241}Am can vary from a few mg/ℓ to several g/ℓ.

Safeguards interests and process monitoring and control consideration require a plutonium assay method that is accurate and timely. Transmission-corrected gamma-ray counting is a nondestructive assay (NDA) method that satisfies these criteria. Briefly, this method requires the measurement of a characteristic isotope gamma-ray rate, R, from a sample and

the transmission coefficient, T, of an external gamma-ray source through the sample at the same energy. The isotope mass, M, present in the sample is given by

$$(1) \qquad M = \left(\frac{R}{K} \right) CF \quad ,$$

where K is a calibration constant and CF is a sample self-absorption correction factor that is a function of T.[1-3]

An instrument using this method has been developed and installed in the Plutonium Processing Facility at LASL. This paper discusses the hardware configuration, the operational method, and the instrument evaluation plan. The discussion of the evaluation plan includes calibration and instrument reliability results and a detailed comparison of the instrument assay results with the results obtained from the analytical chemistry laboratory.

INSTRUMENT DESCRIPTION AND OPERATION

The solution assay instrument (SAI) is a minicomputer-based system with several automated peripherals. The hardware and peripherals have been designed and applied in such a manner that operator intervention is minimized; when intervention is required, it is convenient. There are six major hardware components to the instrument: (1) a germanium detector and sample holder, (2) NIM electronics, (3) a minicomputer, (4) an operator console, (5) a mobile maintenance-and-graphics cart, and (6) a digital electronic balance. The system assays the ^{239}Pu sample concentration by a transmission corrected count of the 414-keV gamma ray from ^{239}Pu.

The sample chamber and germanium detector are shown in Fig. 1. The sample chamber holds the sample vial, which is a right-circular cylinder with a volume of 25 ml. With the sample chamber closed, the vial is surrounded by 5 cm of lead shielding. The transmission source, a plutonium metal disk, is fixed onto a rotating tungsten shutter located above the sample vial. The shutter is operated pneumatically under computer control. Gamma rays from the transmission source and the sample vial are viewed by the detector through a .75-cm tungsten filter and the glovebox floor. The detector has a resolution of 1.7 keV at 414 keV.

Pulses from the detector are processed by standard high resolution gamma-ray spectroscopy NIM electronics, including (1) an amplifier with an internal pulse pile-up rejector, and (2) a two-point energy stabilized analog-to-digital converter.

A Data General-compatible, 16-bit minicomputer is the computational and control tool used in the instrument. The

computer chassis contains the CPU, 32K words of core memory, and the serial communications board. Typical instruction times are 1200 ns; communication data rates are at 300 baud. Control functions and measurements carried out by the computer are actuated from the operator console. A 16-key push-pad is the interface between the instrument, the peripherals, and the operator.

The mobile maintenance-and-graphics cart has a paper-tape reader to load the instrument code or diagnostic programs. The graphics display, similar to a standard multichannel analyzer (MCA) display, allows the user to view the pulse height distribution and to enter specified regions of interest around the photopeaks required by the analysis software. The cart is attached to the system only during the set-up and maintenance periods.

To give the SAI the capability of reporting assay results as g ^{239}Pu/g sample, an electronic balance is located inside the glovebox. Because the glovebox atmosphere is corrosive, the balance digital and analog electronics are external to the glovebox. The balance reading is automatically transmitted to the computer memory by depressing a key on the operator console.
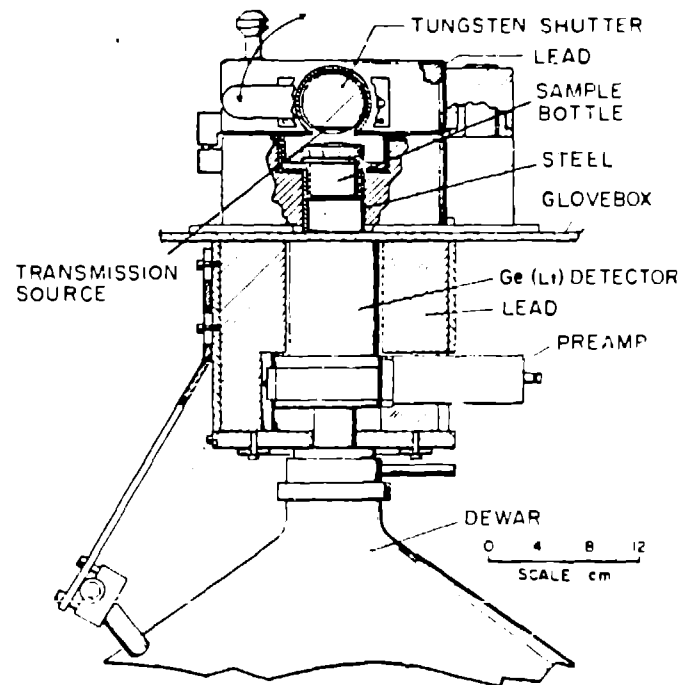
Fig. 1.
Sample chamber assembly
and germanium detector.

To determine the ^{239}Pu mass in a sample aliquot, measurements are required of: (1) the transmission source, (2) the background, (3) the sample, and (4) the transmission source plus the sample. The first measurement determines the unattenuated 414-keV gamma-ray rate from the transmission source. The background measurement records the level of ambient background and is used to correct the rate of the assay peak. These two measurements are performed once or twice a day, depending on glovebox conditions. The last two measurements are performed for each sample and are combined by the system so that no intermediate operator intervention is required.

The third measurement determines the 414-keV gamma-ray rate from the sample; i.e., R in Eq. 1. The fourth measurement sums this rate and the attenuated transmission source signal. The difference between this rate and that measured in the third step, when divided by the 414-keV gamma-ray rate measured in step one, determines the transmission coefficient, T, which in turn allows the sample self-attenuation correction factor, CF, to be calculated (see Eq. 1).

All measurement sequences are initiated by pressing an appropriately labeled key on the operator console. The operator is then led through the required assay steps by a series of computer programmed prompts that appear on the system terminal. When complete, the assay results are printed on the terminal, expressed as g ^{239}Pu/g sample and the associated uncertainty.

RESULTS

The SAI was calibrated using six plutonium standards prepared by the LASL analytical chemistry laboratory. The plutonium concentrations ranged from approximately 0.5 g/ℓ to 300 g/ℓ. The absolute error on the plutonium concentrations was estimated at 0.2%. The SAI calibration results are summarized in Fig. 2, where the percent deviation of the individual calibration constants from the weighted average is plotted versus plutonium concentration. These data show that the calibration constant is concentration independent over a wide range of sample self-attenuation; i.e., the functional dependence of the self-attenuation correction factor upon the measured transmission is correct.

The calculated statistical precision of a single sample assay as a function of plutonium concentration was consistent with the measured precision obtained from repeated assays of the standards, and was found to be consistent. Figure 3 shows the precision obtained for concentrated solutions with a routine 1000-s assay (a 500-s count for the sample and a 500-s

count for the sample plus the transmission source). The decrease in precision at higher concentrations is due to an increased uncertainty in the measured transmission. At lower concentrations the precision varies from 4% at 0.5 g Pu/ℓ to 1% at 40 g Pu/ℓ for a routine 2000-s assay (a 1900-s count for the sample and a 100-s count for the sample plus the transmission source). This decreasing precision at lower concentrations is due to an increased uncertainty in the sample count.

As part of the routine measurement control program, one or more standards are assayed per day as unknowns. The assay value must fall within one standard deviation of the standard value before the SAI may be used for plant sample assay. Figure 4 summarizes these measurement control data for a time period of approximately one month. The eight standards used varied in concentration from 5 to 260 g Pu/ℓ. The average assayed value showed a slight positive bias relative to the standard value, 0.3%, with a standard deviation of 0.7%. The latter is consistent with the measurement precision and indicates reasonably long-term instrument stability.

The SAI accuracy for plant samples was evaluated by comparison with three analytical chemistry methods: isotopic dilution mass spectrometry (IDMS), radiochemical alpha particle counting, and coulometric titration. Samples with plutonium concentrations greater than 40 g/ℓ were chemically assayed by coulometric titration. Those with plutonium concentrations less than 40 g/ℓ were assayed by either IDMS or by alpha particle counting. The results of this comparison are summarized in Fig. 5 where the assay differences in percent are plotted versus plutonium concentration. The sample error bars reflect only the SAI uncertainty and include contributions due to statistical precision, sample volume, calibration, and isotopic abundance uncertainties.

The IDMS method has a routine accuracy of 0.2 to 1%, the major fraction of which is due to the complex sample preparation procedures. The SAI·IDMS comparison is shown in Fig. 5a. A small positive bias is observed, 0.24%, with a standard deviation of 1.16%. This standard deviation is consistent with the calculated SAI uncertainty for this plutonium concentration range. The bias is consistent with that observed for the measurement control data discussed above and may indicate a small error in the SAI calibration constant.

Due to the expense and time required for the IDMS method, it is rarely used to assay plant samples. Plant samples were routinely assayed by the alpha particle counting method before installation of the SAI. The accuracy for this method is limited to approximately 5 to 10%, due to sample preparation difficulties.
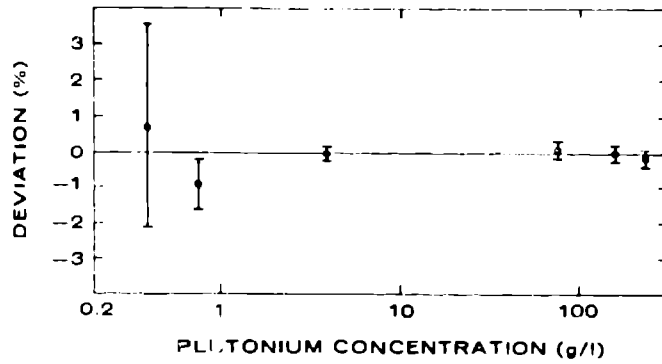
Fig. 2.
Calibration constant deviation as a function
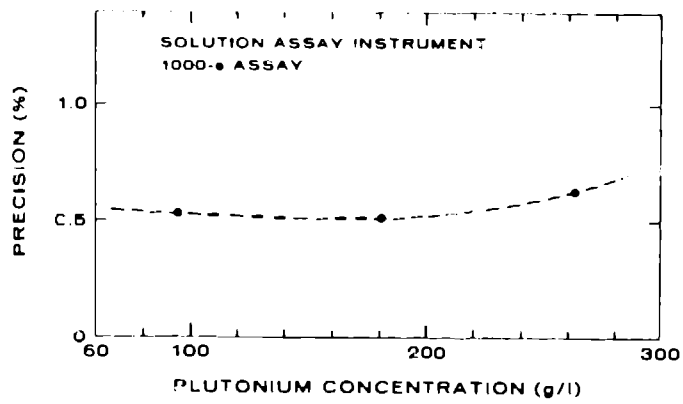of plutonium concentration.

Fig. 3.
Assay precision as a function of plutonium
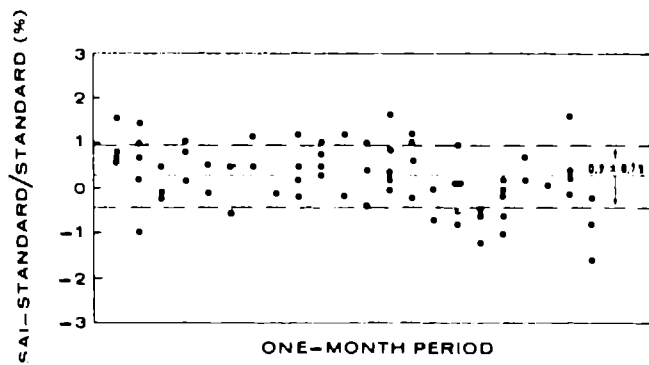concentration for a 1000-s count.

Fig. 4.
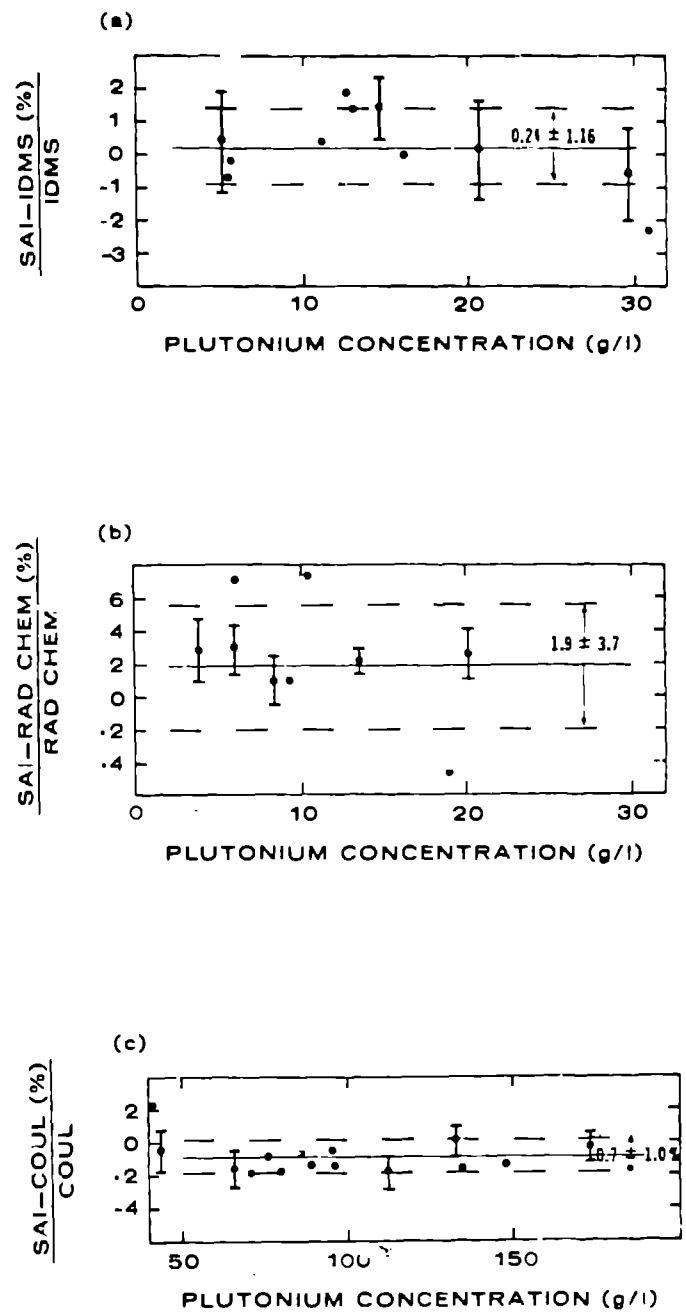Instrument stability as a function of time.

Fig. 5.
Comparison of SAI assay results
with analytical chemistry results.

The SAI-radiochemical comparison is shown in Fig. 5b. The bias observed here is approximately 2% with a standard deviation of 3.7%. A comparison of these results with those presented in Fig. 5a shows that the SAI provided increased accuracy for routine plant sample assay.

The results of the SAI-coulometric titration comparison for concentrated plutonium solutions are summarized in Fig. 5c. These data indicate a bias of -0.7% and a standard deviation of 1.0%, which is consistent with the SAI accuracy. The SAI measurement control data show no bias for the standards assayed during the same period that the plant samples were assayed. The titration technique has a routine accuracy of 0.2 to 0.5%. This accuracy depends on proper correction for iron content. An inadequate correction results in a negative bias.

DISCUSSION

The SAI has proven to be a reliable and accurate instrument for providing timely assays of plutonium solutions over a wide concentration range. This instrument is being routinely used by process personnel who have no detailed knowledge of the instrument. The accuracy and precision of the SAI assay results compare favorably with analytical chemistry results. Furthermore, use of the instrument does not require complex sample preparation. These qualities suggest that the SAI can be a valuable tool for a materials management program and process control in a plutonium processing facility.

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