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DEVELOPMENT OF A PLUTONIUM SOLUTION-ASSAY INSTRUMENT WITH ISOTOPIC CAPABILITY*

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ABSTRACT

A new generation of solution-assay instrument has been developed to satisfy all the assay requirements of an aqueous plutonium-recovery operation. The assay is based on a transmission-corrected passive assay technique. We have demonstrated that the system can cover a concentration range of 0.5–300 g/ℓ with simultaneous isotopic determination. The system can be used to assay input and eluate streams of the recovery operation. The system can be modified to measure low-concentration effluent solutions from the recovery operation covering 0.01–40 g/ℓ. The same system has also been modified to assay plutonium solutions enriched in ^{242}Pu.

INTRODUCTION

Plutonium solution-assay instruments are needed to determine the plutonium concentration for accountability purposes. This work reports our development of a new system with a wide dynamic range (0.01–300 g/ℓ) and determines both plutonium concentration and isotopic distribution simultaneously for in-plant application. This type of system meets all the assay requirements of an aqueous recovery operation.

PROBLEM

The request for developing such a system came from the Los Alamos Plutonium Facility (LAPF). In this facility, there is a plutonium operation to recover the plutonium from scrap and waste. The major aqueous process used to recover and purify plutonium at LAPF is anion exchange in nitric acid. This process is well suited for purifying plutonium, as Pu(IV) is more strongly sorbed than any other ion, and few other metal ions show even moderate sorption from nitric acid. A continuous-feeding dissolver has been installed to speed up the dissolution, and because it is not a batch process, the concentration and isotopic distribution of the solution is unknown. This dissolved plutonium solution (10–200 g/ℓ), loads on the anion exchange column, the eluate stream contains the purified plutonium with concentrations ranging from 5–50 g/ℓ, and the effluent contains most of the impurities with traces of plutonium (~10 ppm or more, depending on the success of the separation).

Ten years ago, a plutonium solution-assay instrument (SAI)[1,2] was built and installed at LAPF. This instrument has served its purpose of providing a solution-concentration-determination capability in the process line. However, we have also found some operational problems:

1. Some of the solutions at Los Alamos are found to contain abnormally large amounts of ^{237}Np. In the previous SAI, assays of plutonium solutions contaminated with ^{237}Np and ^{233}Pa may have caused excessive bias because of the presence of the 415.76-keV gamma ray from ^{233}Pa.
2. Mechanical problems. Some of the mechanical parts, which worked well in the beginning, started to develop problems in the hostile acid environment of the glove box after several years. The tungsten shutter, despite the fact that it is gold plated, has a tendency to corrode in the acid atmosphere and after a period of time has difficulty in rotating and shuttering the transmission source.

To eliminate these problems, a new generation of SAI has been developed to determine the plutonium concentrations and isotopic distribution simultaneously. The instrument is designed to assay a wide range of solutions (0.5–300 g/ℓ) mixed with impurities in the plutonium-recovery operation. They have been used to assay both the input solutions to the anion exchange separation as well as the eluate stream. A variation of

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the SAI has also been developed to assay low-level solutions from the effluent streams of the process, covering a concentration range of 0.01–40 g/ℓ. In response to the facility request, one of the SAIs has been extended to assay plutonium solutions enriched with ^{242}Pu.

HARDWARE

The hardware of the new SAI is shown schematically in Fig. 1. The hardware that resides in the glove box consists of three segments. The top segment contains the transmission source, shutter, and shutter motor. The ^{75}Se transmission is in a plastic holder in a tungsten shield and can be changed from the top without opening the segment. The tungsten shutter is driven by a dc gear motor; the direction of the shutter is controlled by a reversing actuator. This segment of the SAI is filled with clean air to prevent acid corrosion. At the top of the segment, there are three lights showing the shutter position, assay chamber position, and assay-in-progress status indicator. The new design eliminates the problem with the shutter, as in the previous design.

The second segment is mainly a stainless-steel housing filled with lead to shield the detector from background radiations; the center of this segment is a tungsten collimator for the transmission source. The bottom segment is lead shielding encapsulated in stainless steel; in the center is a plastic well to hold the

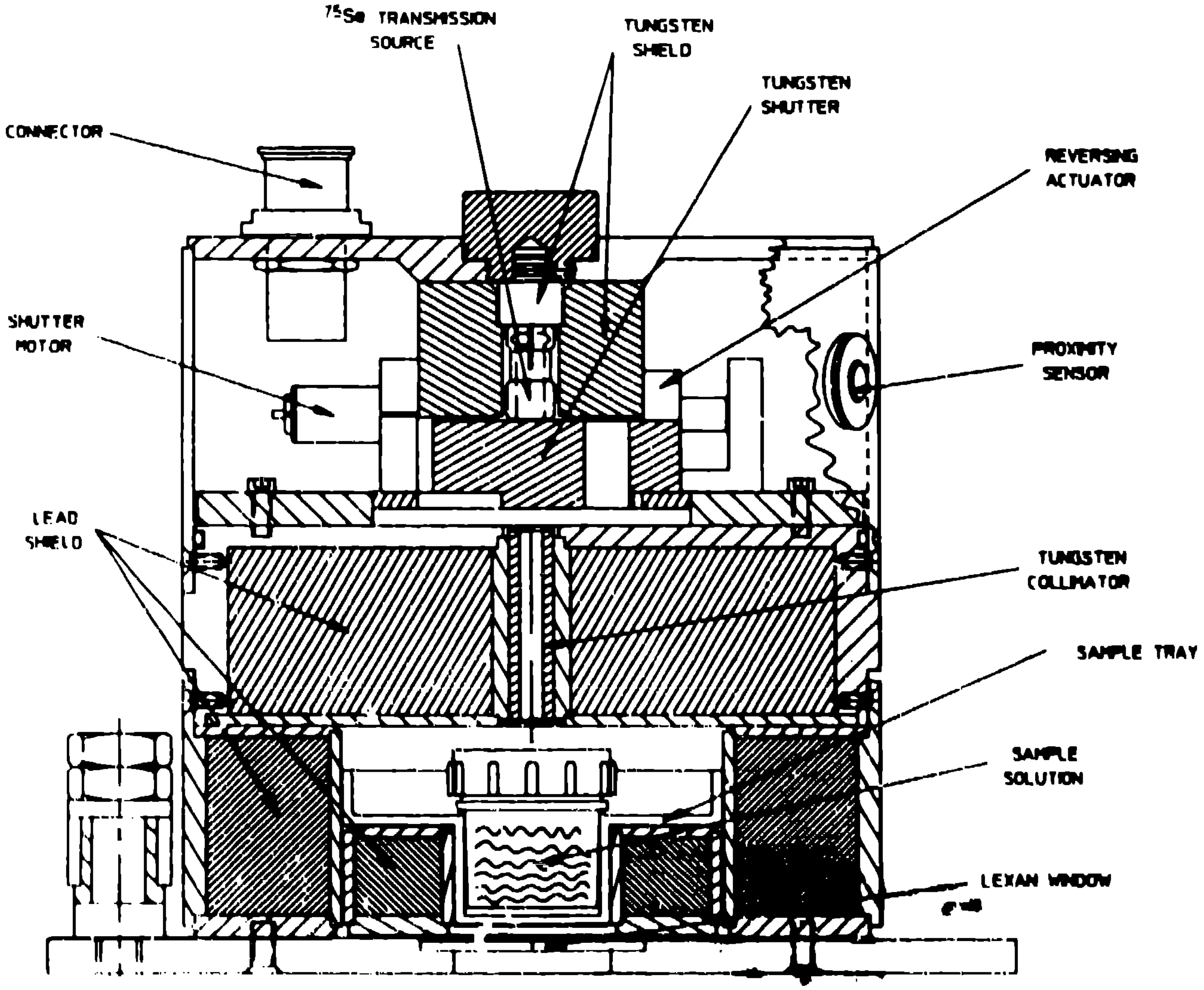

Fig. 1 Sample chamber residing in the glove box. The top portion contains the transmission source, tungsten shutter, and shutter motor. The bottom portion contains the solution sample vial.

sample-solution vial during assay. This segment is hinged and can be opened for sample loading and unloading.

The detector is located outside the glove box on a detector stand, looking at the sample through the glovebox wall, so no modification to the glove box is necessary. The detector is a low-energy photon, spectrometer type with an active volume of 36-mm diam by 15-mm depth. The size of the detector is selected so that the detector surface area is the same as the sample surface area. To perform isotopic analysis, the detector resolution should be good, with little or no higher energy tailing. The front surface of the detector is located $\sim$13 cm below the solution sample. The volume of the sample is 25 mℓ. A picture of the setup is shown in Fig. 2.

ANALYSIS

To eliminate bias problems resulting from the presence of ^{237}Np and ^{233}Pa, gamma peaks in the spectrum are fitted with a response-function technique.[3] Protactinium-233 emits a 415.76-keV gamma ray that interferes with the 413.71-keV gamma ray of ^{239}Pu, which is the main peak used for assay in the region-of-interest method of peak-area determination. This is resolved by peak fitting of overlapping peaks; peak fitting also allows for the presence of a potential pileup peak at 2 x 208 or 416 keV, although the peak shape of a pileup peak is different from a regular photopeak. Because the fitting technique can tolerate the pileup peak, the 0.76-mm-thick tungsten filter, which was used in the previous system, is not necessary; therefore, the 129.29 keV can also be used to assay ^{239}Pu, which improves the precision at low concentrations. For assaying of ^{239}Pu, 129.29-, 345.01-, and 413.72-keV gamma rays are used as a signature.

Fig. 2. Sample chamber in the glove box and the detector outside the glove box.

To make quantitative assays at these energies, the sample self-absorption correction is necessary when assaying over a wide concentration range and a variety of solution types. The transmission source selected is ^{75}Se: the transmissions are measured at 136.00, 279.53, and 400.65 keV. These transmissions are interpolated to other energies by means of quadratic fits in the Ln[-Ln(Transmission)] vs Ln(Energy) space. The interpolation has been discussed in previous work.[4] The attenuation correction factor is calculated using a one-dimension model by numerical integration.[5]

To determine the plutonium isotopic distribution in a relatively short time (<500 s), the MGA2 technique[6] is used. This technique utilizes the gamma-ray information from 60–208 keV to determine the plutonium isotopic distribution.

SYSTEM FOR LOW-CONCENTRATION SOLUTION

A variation of SAI tailored for a low-concentration plutonium solution has been developed so that the effluent stream from the anion exchange will contain most of the impurities with a trace of plutonium. The major interferences arise from the ^{241}Am and ^{237}U present in the solution. Americium emits a strong 59.54-keV gamma ray, which needs to be filtered so that it does not dominate the count rate. The ^{237}U emits a 208-keV gamma ray and because of the 6.8-day half-life, is also quite intense; the pileup peak of 2 x 208 at 416 keV interferes with the 413.71-keV gamma ray, which is the main peak used in the ^{239}Pu assay. In addition, the plutonium concentrations in these solutions are relatively low.

To assay these types of solutions, the efficiency of detecting the weak signals from the relatively low plutonium concentration needs to be increased. This is accomplished by using a larger, more efficient detector viewing a greater volume of sample compared to the SAI system. A coaxial detector (50-mm diam by 40-mm depth) with an efficiency of 20% (at 1.33 MeV) and a resolution of 850 eV at 122 keV is used in such a system; the sample volume has been increased from 25 $m\ell$ to 60 $m\ell$; the distance between the solution and the detector surface has been reduced from ~13 cm to ~6 cm. The size of the detector is again selected so that the surface area is the same as the sample-solution surface area. The SAI hardware must be modified to accommodate the larger vial, which is a minor modification. Plutonium isotopic distribution in these waste solutions can also be determined with this detector, although only with the relatively higher concentration samples. In these cases, the ^{239}Pu weight fraction must be entered. The sensitivity of the system is ~10 mg/ℓ in a 2000-s assay.

SYSTEM FOR PLUTONIUM SOLUTIONS ENRICHED IN ^{242}Pu

Another variation of the SAI tailored for plutonium solutions enriched in ^{242}Pu has also been developed. LAPF is recovering plutonium enriched in ^{242}Pu also by the anion-exchange process and generating solutions that need to be assayed.

Plutonium-242 emits very few gamma rays that can be used as an assay signature, so gamma rays from other plutonium isotopes must be used for assay. Table I shows the typical isotopic distributions of the two types of the ^{242}Pu material.

From this table, it is obvious that because of the relatively low percentage of ^{239}Pu in these materials, it is not the appropriate isotope to use for assay; however, the weight percents of both ^{238}Pu and ^{241}Pu are reasonably abundant and can be used. Plutonium-238 has a gamma peak at 152.68 keV, and ^{241}Pu has a gamma peak at 148.57 keV, both of which can be used for assay purposes.

We have modified an SAI system to assay both routine low-burnup plutonium solutions and solutions enriched in ^{242}Pu. To assay for ^{239}Pu, the system is calibrated at 129, 345, and 414 keV. These calibration constants are used to interpolate the calibration constants at 148 and 152 keV. Therefore, the same system calibrated for ^{239}Pu can also be used to assay ^{238}Pu and ^{241}Pu without an additional calibration effort.

CONCLUSION

The SAI is sufficiently versatile that it can be adapted to assay a wide range of concentrations and various sample types. We have demonstrated that the SAI type of instrument can meet all assay requirements of an aqueous recovery operation.

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TABLE I. Typical Isotopic Distribution of Two Types of ^{242}Pu (in wt %)

	^{238}Pu (wt %)	^{239}Pu (wt %)	^{240}Pu (wt %)	^{241}Pu (wt %)	^{242}Pu (wt %)
Type 1	0.4500	0.4364	2.401	0.9185	95.794
Type 2	1.035	1.389	10.34	3.214	83.999

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