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AMERICIUM ASSAY INSTRUMENT

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ABSTRACT

A simple in-line americium assay instrument (AAI) was installed at an americium recovery process area at Los Alamos Scientific Laboratory (LASL) for use in process development and for providing process control information. The AAI counts 59.5-keV ^{241}Am gamma rays, using a NaI(Tl) detector and Eberline SAM II electronics. It has a useful range of 3×10^{-5} to 10 g Am/ℓ and does not suffer from plutonium interference. Comparative analyses of samples assayed in the AAI and samples assayed by the LASL Analytical Laboratory show a combined relative standard deviation of 14%.

Nondestructive assay (NDA) methods have been applied to a wide range of special nuclear material (SNM) control problems. Many of the NDA instruments have proven useful as process control monitors, providing more timely determination of material flow than that available from analytical chemistry laboratories. The safeguards application of these instruments generally requires a reasonably sophisticated design and a high degree of measurement accuracy. For process control, however, these requirements can often be less restrictive, especially when quick installation is required.

An americium recovery process, based on ion exchange and selective precipitation, recently began operation at the Los Alamos Scientific Laboratory (LASL) Plutonium Processing Facility. An in-line NDA instrument, capable of measuring the americium concentration of process solutions, was designed at LASL to meet the following criteria for process development:

(1) an ^{241}Am analysis range of 1×10^{-3} to 5 g Am/ℓ,
(2) an analysis time on the order of minutes,
(3) analysis accuracy of ±25% relative,
(4) analysis of americium in solutions having plutonium-to-americium ratios as high as 100:1,
(5) operation in a glovebox with kilogram quantities of plutonium and gram quantities of americium within a 3-m radius,
(6) installation that does not require cutting the glovebox structure, and
(7) simple operation and calibration.

This paper describes the LASL americium assay instrument (AAI), which applies a straightforward NDA method to satisfy these criteria.

The AAI was designed to measure the very intense, relatively interference-free, ^{241}Am gamma ray at 59.5 keV. This gamma ray is sufficiently energetic to penetrate the glovebox floor with only 40% attenuation. Because the solutions measured were of relatively low concentration (plutonium + americium less than 15 g/ℓ), no correction was necessary for sample self-absorption.

The AAI hardware is shown in Fig. 1. The gamma rays are detected by a 5- x 5-cm sodium iodide detector. The detector, photomultiplier tube, and preamplifier are encased in a 12-mm-thick lead collimator and mounted under the glovebox. Signals from the detector are processed by an Eberline SAM II electronics package, which contains the required high-voltage power supply, amplifier, single-channel analyzer, scaler, and timer. The detector high voltage is stabilized using a reference signal that is generated by an alpha source in the sodium iodide detector.

The sample holder, shown above the collimated sodium iodide detector in Fig. 1, was designed for analyzing low-concentration americium solutions. After alignment with the sodium iodide detector, the sample holder was bonded to the glovebox floor with a silicone adhesive. The holder, machined from one-half of a lead brick, shields the detector from background radiation while precisely positioning the sample bottle close to the detector.

For analyzing higher concentration solutions, a collimator was placed inside the sample holder. The collimator lengthens the distance between the sample and the detector, and restricts the gamma rays counted to those passing through a 3.8-mm-diam opening. This adapter reduced the americium count rate by a factor of 180. The lower curve in Fig. 2 shows the pulse-height distribution measured for an americium sample with a very low plutonium-to-americium concentration ratio of 1:50, or 0.02. On the basis of these data, the single-channel analyzer window was

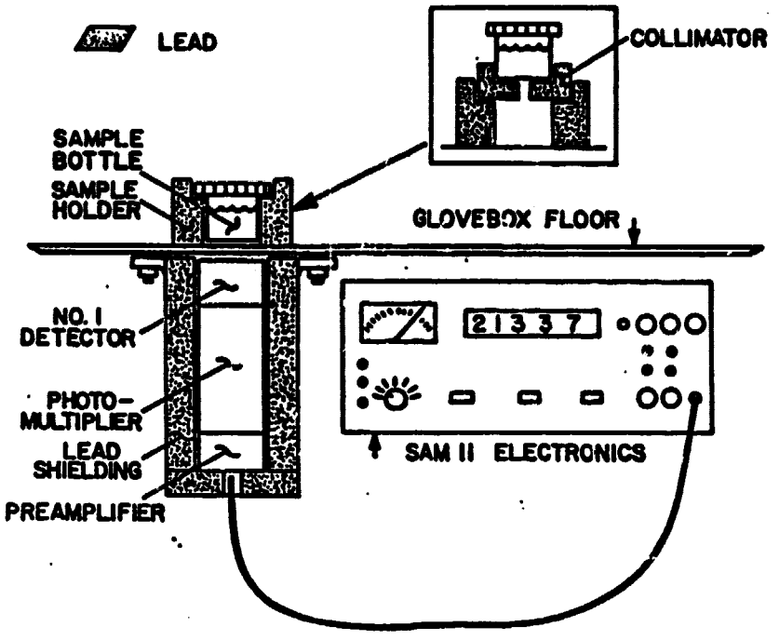

Fig. 1.
AAI components.

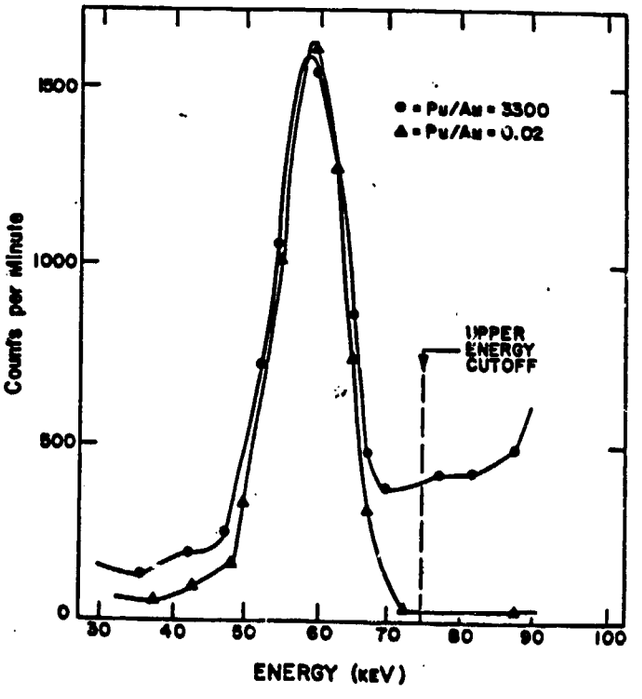

Fig. 2.
Pulse height distribution.

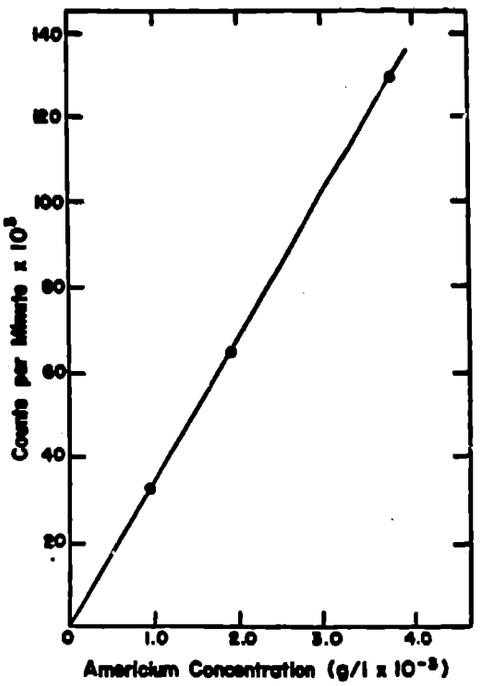

Fig. 3.
Uncollimated calibration curve.

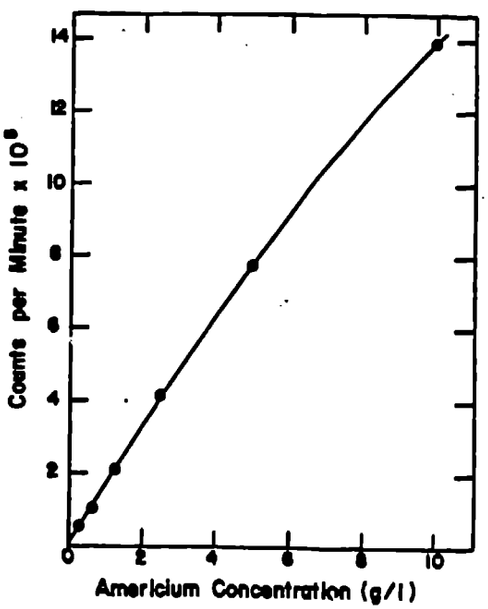

Fig. 4.
Collimated calibration curve.

set to accept pulses whose amplitudes corresponded to gamma rays with energies of 35 to 75 keV.

Plutonium contamination in the samples affects the accuracy of the americium assay by three different mechanisms: (1) introduction of plutonium gamma rays into the assay energy region, (2) introduction of a Compton continuum under the assay region, and (3) increase in the pulse pile-up and deadtime rate.

Plutonium gamma-ray activity in the 35- to 75-keV region was calculated assuming a plutonium isotopic composition characteristic of FFTF fuel. These plutonium gamma peaks add about 0.2% to the ^{241}Am peak area for a 100:1 plutonium-to-americium mixture, and about 2% for a 1000:1 plutonium-to-americium mixture.

Figure 2 shows the influence of the Compton effect upon the pulse-height distribution for a sample with a plutonium-to-americium ratio of 3300:1. For a solution with a plutonium-to-americium ratio of 1000:1, the Compton continuum biases the americium assay about 2% high.

The increased pulse pile-up and deadtime rates biased the assay low. Deadtime losses were less than 1% for americium count rates up to 100 000 counts/min. However, plutonium gamma rays outside of the counting window contributed to pulse pile-up. Measurements on standards having varying plutonium-to-americium ratios indicate that pile-up from plutonium gamma rays becomes significant when the plutonium concentration exceeds 10 g/ℓ.

The AAI was calibrated with a set of standards whose americium concentrations varied from 1×10^{-5} to 10 g/ℓ. The results for the uncollimated and collimated counting geometries are summarized in Figs. 3 and 4, respectively, where the count rates of the standards are plotted versus americium concentration. In the uncollimated geometry, the calibration curve slope is constant for concentrations less than 5×10^{-3} g Am/ℓ. Similarly, the calibration curve slope is constant in the collimated geometry for concentrations less than 1.5 g Am/ℓ. For higher concentrations, the curve slope decreases, due to pile-up and deadtime losses.

To determine the AAI's accuracy on routine plant samples, the AAI assay was compared with an analytical laboratory assay based on gamma-ray counting. The routine accuracy of the latter is approximately ±1%. The results are summarized in Table I.

The AAI analysis consisted of a 60-s background count and a 60-s sample count. The uncertainty of the counting statistics associated with the measurements is less than 3%. Comparison between the two assay methods shows that the AAI results are

biased by 1.4%, with an average deviation of ±13.5%. These results are well within the design criteria.

In summary, the AAI is a simple instrument to assemble, install, calibrate, and operate. Using a 1-min background and 1-min sample counting time, solutions varying from 1 x 10^{-3} to 10 g Am/ℓ can be analyzed with an accuracy of better than 14%. Plutonium interference is insignificant if the plutonium concentration in the sample remains below 10 g/ℓ and the plutonium-to-americium concentration ratio remains below 100:1. At LASL the AAI met or exceeded all the initial design requirements. Process personnel used the AAI to obtain the rapid americium analyses needed during development of the americium recovery process.

TABLE I

AAI AND ANALYTICAL LAB ANALYSES OF ^{241}Am SAMPLES

Sample Number	AAI (g Am/ℓ)	Lab (g Am/ℓ)	Percent Difference[a]
LAF25	0.18	0.26	-31
LAF26	0.25	0.23	9
LAF27	0.27	0.44	-39
LAF28	0.51	0.47	9
LAF29	0.92	0.81	14
LAF30	0.66	0.61	8
LAF31	0.36	0.33	9
LAF32	0.35	0.27	30
LAF33	0.55	0.59	-5
LAF34	0.39	0.40	-3
LAF35	0.24	0.24	0
LAF36	0.24	0.25	-4
LAF37	0.048	0.045	7
LAF38	0.085	0.085	0
LAF39	0.079	0.075	5
LAF40	0.035	0.036	-3
LAF41	0.041	0.038	8
LAF42	0.031	0.029	7
LAF43	0.081	0.076	7
LAF44	0.057	0.055	4
LAF45	0.084	0.082	2
LAF46	0.61	0.61	0
LAF47	0.27	0.29	-7
LAF48	0.60	0.61	-2
LAF49	0.74	0.68	9

[a] $\left[\frac{\text{AAI - LAB}}{\text{LAB}}\right] \times 100$