MEASUREMENT OF PLUTONIUM AND AMERICIUM IN MOLTEN SALT RESIDUES *

by

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ABSTRACT

The measurement of plutonium and americium in molten salt residues using a segmented gamma-ray scanning device is described. This system was calibrated using artificially fabricated as well as process generated samples. All samples were calorimetered and the americium to plutonium content of the samples determined by gamma-ray spectroscopy. For the nine samples calorimetered thus far, no significant biases are present in the comparison of the segmented gamma-ray assay and the calorimetric assay. Estimated errors are of the 10 percent and is dependent on the americium to plutonium ratio determination.

KEYWORDS: Waste assay, plutonium, americium, nuclear safeguards, nondestructive assay.

INTRODUCTION

A segmented gamma-ray scanning device has been installed at Rocky Flats for measurement of Waste residues in small, less than one gallon, containers. Initially, the system has been programmed to assay crushed molten salt residues for plutonium and americium. The americium assay is required to account for the decay of the isotope 241-Pu. The salt residues are generated in the plutonium recovery process by a molten extraction procedure. The spent chloride salt media from the extraction contains all of the americium and some of the plutonium present in the plutonium feed material. Plutonium content of the salts ranges up to 400 grams and americium content to 30 grams. Typically, the ratio of americium to plutonium is 5- to 10 percent. Matrix weights are of the order of 2 kilograms.

MEASUREMENT PROCEDURE

The measured salts are doubly contained in stainless steel cans: A Volrath 8801 can inside a volrath 8802 can. The outside dimensions of the sample cylinder are approximately 4.25 inches in diameter and 5.5 inches tall. A commercially available counting system, Canberra Industries Model 2220C, (Ref. 1) is used to assay the salts. The sample can is divided into five segments and each segment is counted for 200 seconds. The plutonium is measured using the area of the 414-keV peak; the americium is measured using the area of the 662-keV peak. We also monitor the intensity of the 393-keV peak due to 239-Pu.

Gamma ray attenuation corrections are made for all of these gamma rays using the 400 keV gamma-ray peak from an external 75-SE transmission source. The relative attenuation correction for the 400 keV gamma ray to that occuring at the 662-keV americium gamma ray was measured by recording the transmission of gamma rays from the 72-SE source and an external 137-CS source, energy 662 keV, through a molten salt cake containing no radioactive contaminants. The measured ratio of the mass attenuation coefficients at 662 keV is 0.78.

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DISCLAIMER

Figure 1 shows a gamma-ray spectrum of the 400 keV energy region from a sample containing approximately 10 percent americium relative to plutonium. High count rates from the sample required that we incorporate pulse pile-up rejector electronics into the system. This reduced the high energy tailing on the accumulated peaks and the amount of coincident summing in the crystal of the 208 keV peak from the decay of 237-U and 241-Am. This latter peak, 416 keV, is now seen as only a slight "bump" on the high energy base of the 414-keV peak. The pile-up rejector electronics has allowed us to maintain a simple background subtraction procedure for the areas of the peaks. By monitoring the area of the 393-keV peal relative to the area of the 414 keV peak, we have ascertained that this model of area determination is adequate for americium levels below 15 percent relative to plutonium. The ratio of 393-keV assay to that obtained from the 414-keV assay range from 0.97 to 1.03 with the uncertainty of each determination being of the order of 4 percent or less.

CALIBRATION

The system has been calibrated for plutonium by fabricating salt standards containing 100-, 200-, and 400 grams of plutonium. These standards contain no americium and have been calibrated by calorimetric techniques. Agreement of the gamma-scan values with those obtained from calorimetry were less than 3 percent. System reproducibility for these samples was +/- 2 percent.

The americium calibration was obtained by preparing two salt samples into which weighed quantities of americium of 6 and 29 grams, respectively, were added. Both of these samples were contaminated with about 25 grams each of plutonium. This was verified by both the calorimeter and the gamma-ray spectra. It was these samples that called our attention to the interferences present at 416 keV due to pile-up of the 208 keV gamma-ray and at 419 keV due to the americium in the samples. To investigate the extent of these interferences three samples were prepared having approximately 5-, 10-, and 20 percent americium relative to plutonium. Again, contamination of the samples occurred as indicated.

By comparison of the calorimetric assay with the assigned weights, however, the agreement between the calorimetric assay and the segmented gamma assay was within 5 percent. The calorimetric determination in this latter case was calculated using the americium to plutonium ratio determined by gamma-ray spectroscopy using the 125 keV peak from americium and the 129 keV peak from plutonium. Five spectra are summed, one spectrum from each segment, and the ratio determined from the summed spectrum.

Calibration of the system was also undertaken using samples selected from inventory. The Department of Energy, Albuquerque Operations Office, Division of Safeguards and Security (DOE/ALO-DSS) requested that the molten salt waste category undergo an inventory verification study for their most recent audit. Nine samples were chosen. Each sample was calorimetered and the americium/plutonium ratio determined as above. All of the packages were chosen on a highly selective basis because of the limited calibration range, less than 3 watts, of the calorimeters used for the study. In addition, the selection involved the handling of a minimum number of drum storage containers to minimize the health hazards to the operating personnel.

RESULTS AND DISCUSSION

Figure 2 shows the results of the comparison of the plutonium values obtained by calorimetry versus those measured by the segmented gamma-ray system. The points represented

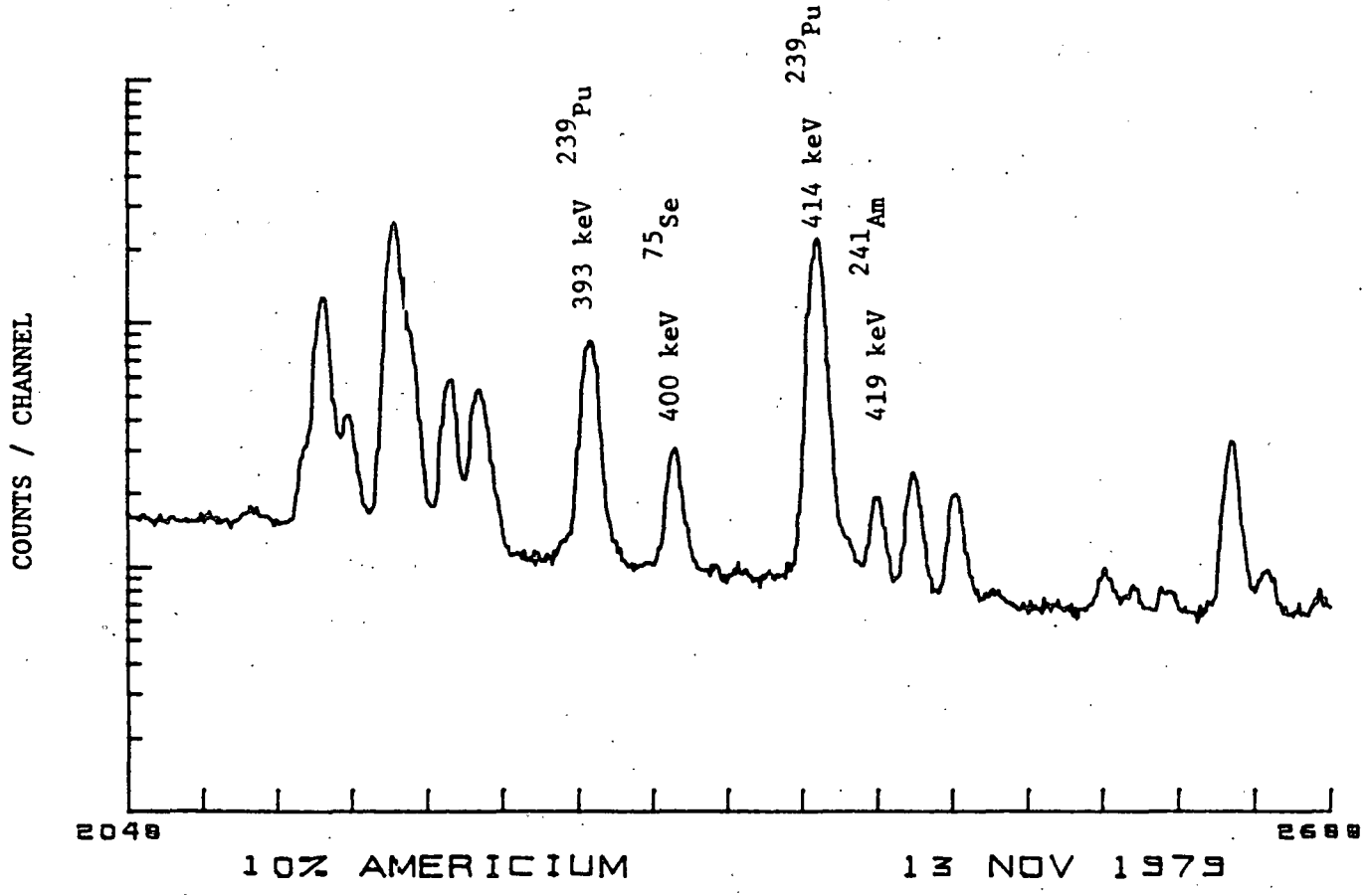

Figure 1 Spectrum of the 400 keV energy region used for segmented gamma-ray assay. Peaks used in the analysis are the 393- and 414-keV peaks from the decay of 239-Pu.

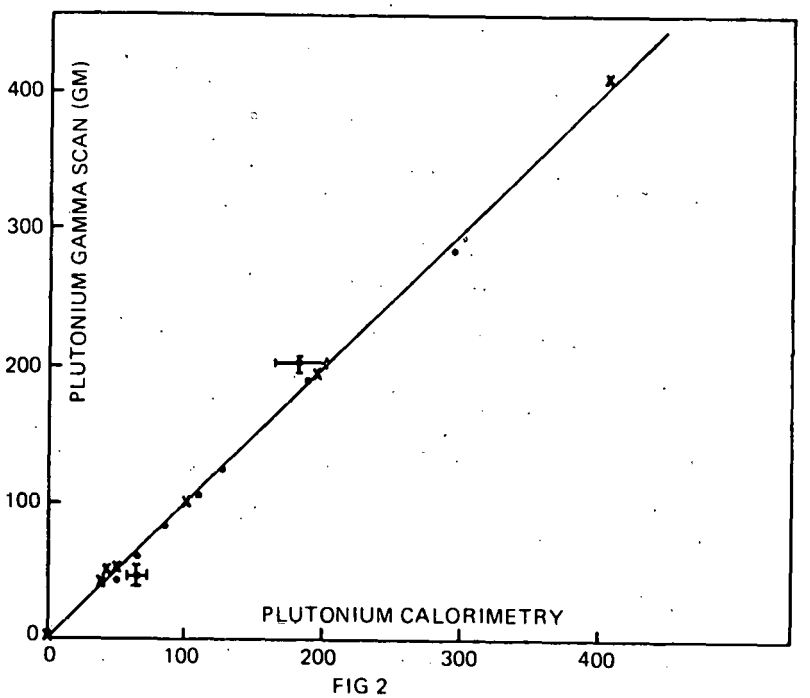

FIG 2

Comparion of plutonium values obtained by calorimetry and segmented gamma scan. Dots represent values from inventory samples. x's represent values from artificially made salts. See text.

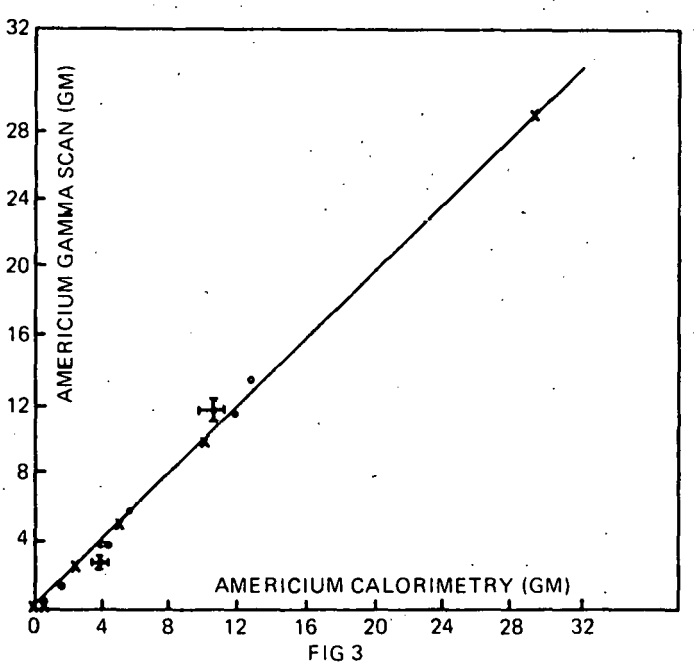

FIG 3

Comparison of americium values obtained by calorimetry and segmented gamma scan. Dots represent values from inventory samples. x's represent values from artificially made salts.

by x's are the points measured for the prepared standards. The dots represent the data for the DOE/ALO-DSS verification samples. Figure 3 shows the corresponding data for the americium values. The straight line indicates the locus of points where the segmented gamma scan values equal the calorimetry values. The predominant error in this technique is that propagated by the uncertainty in the americium/plutonium ratio determination applied the calorimetry values. This error is estimated to be 7- to 10 percent based on comparisons with the same ratio obtained using the 419-keV and 414-keV gamma rays and previous results relative to the verification program (Ref. 2) since the americium contributes 30- to 90 percent of the heat in all of the samples, except the pure plutonium samples, the overall uncertainty in the plutonium determination by calorimetry is then of the order of 3- to 10 percent. Error bars on all the points overlap the line of equality in the figures, except those points with the error bars indicated.

A statistical study of the comparison of the results from the gamma-ray segmented scan with calorimeter results for the nine inventory samples indicate a standard bias for the samples of -5.1 percent with an error of 6.2 percent. For the small number of samples studied, the relative biases are not statistically significant at the 95 percent confidence level. The study of biases is continuing at the present time, four samples per month will be taken from inventory, calorimetered, and counted at least once per week for the month. This process will continue until a statistically significant number of samples are verified and biases, if any, determined.

REFERENCES

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2. John F. Lemming, Francis X. Haas, and Jack Y Jarvis, "Gamma-ray Isotopic Measurements for the Plutonium Inventory Verification Program," MLM-2312 (Aug. 25, 1976)