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**Nuclear Safeguards Progress Report:
July 1974 - June 1975**

December 19, 1975

Research and Development Report

MOUND LABORATORY

Miamisburg, Ohio

operated by

Monsanto

MONSANTO RESEARCH CORPORATION

a subsidiary of Monsanto Company

for the

**UNITED STATES ENERGY RESEARCH
AND DEVELOPMENT ADMINISTRATION**

U. S. Government Contract No. E-33-1-GEN-53

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R. P. Ratay, Editor

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PRINTED IN THE UNITED STATES OF AMERICA

Available from
National Technical Information Service
U. S. Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22161
Price: Printed Copy \$4.00; Microfiche \$2.25

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Foreword

Under the sponsorship of the ERDA Division of Safeguards and Security and the Nuclear Regulatory Commission, Mound Laboratory is responsible for studies to advance nuclear safeguards technology in the public interest.

This report is submitted by W. T. Cave, Director, Nuclear Operations, and R. E. Vallee, Manager of Technology Applications and Development, from contributions prepared by members of the Applied Physics Section: W. H. Smith, Manager, and the Automation Development Section: C. F. Draut, Manager.

Summaries

DEVELOPMENT OF NONDESTRUCTIVE ASSAY TECHNIQUES

Plutonium Isotopic Measurements by Gamma-Ray Spectroscopy

A nondestructive technique for the determination of the atomic ratios of plutonium-238, -240, -241 and americium-241 relative to plutonium-239 using high-resolution gamma-ray spectroscopy has been developed. The results of these techniques are discussed as they apply to various compositions of plutonium-239 material, some of which have been subjected to chemical analysis. (Page 5)

SYSTEMS DEVELOPMENT

Automated Plutonium Assay System

Progress to date on an automated plutonium assay system is described. This system utilizes calorimetry and high-resolution gamma-ray spectroscopy. It also features a computer controlled X,Y,Z sample-handling system as well as automated data acquisition and reduction. (Page 16)

Development of Nondestructive Assay Techniques

PLUTONIUM ISOTOPIC MEASUREMENTS BY GAMMA-RAY SPECTROSCOPY

*F. X. Haas and
J. F. Lemming*

INTRODUCTION

The purpose of this work is to develop nondestructive techniques for the measurement of plutonium isotopic concentrations and americium-241 with a relative standard deviation of at most $\pm 3\%$. The development work described is a continuation of previous work¹ using high-resolution gamma-ray spectroscopy. New peak areas used for analysis as described in this report follow techniques developed in MLM-2186.¹ Constants depending on gamma-ray branching ratio and isotopic half-life are calculated using values obtained from Gunnink and Morrow.²

PROGRESS IN DATA REDUCTION

Two improvements have been made in the area of data reduction. The input subroutine for reading spectral data from the magnetic tape into the program GAUSS V³ was rewritten. This has allowed spectral-analysis time to be reduced by a factor of seven. It has also produced a significant improvement in the IBM-360 turnaround time.

Secondly, the FORTRAN II computer programs, GRPANL, written by R. Gunnink⁴ have been compiled under OS/8 on our PDP-8/L computer system. These programs were chosen to perform spectral analysis because of the use of the dispersed-step background under the peaks. This is considered to be a more realistic choice in background selection than the linear background used by GAUSS V.

Results of using these two programs are compared in other sections of this report. GAUSS V requires a good deal of user time in preparation of the input cards. Hand calculations of input data are required for the background values and slopes for each peak or peak grouping analyzed. The

GRPANL program requires an equal amount of operator time at the console for inputting data for all of the peak groups used in our analyses. However, the GRPANL program and its calls to its subprograms have been modified at Mound for spectra run at a fixed gain of 0.26 keV per channel (routine spectrum acquisition gain for the Inventory Verification Program)¹ to perform a complete analysis including plutonium isotopics with a minimum of operator intervention.

ISOTOPIC CALCULATIONS

New peak pairs other than those reported previously have been investigated for determining the plutonium and americium concentrations. The primary objective of these determinations is to obtain the isotopic ratios in the low-energy region of the spectrum ($E_\gamma < 400$ keV) where the count rate is high. Use of this portion of the spectrum would reduce the data acquisition time and hence increase the number of samples which can be processed in a given time period.

Figure 1 shows a typical spectrum of weapons-grade plutonium material over the energy region 0-1 MeV. Figure 2 is an expanded portion of a spectrum of NBS-SRM 948 over the energy region 100-400 keV. Both spectra were acquired with a 70-cc Ge(Li) detector, with resolution of 1.80 keV at 1.3 MeV and 1.00 keV at 122 keV. For most of the work reported, count rates of 3000 counts/sec were maintained and a 0.030-in. (0.076-cm) cadmium absorber was placed around the detector housing.

In addition some of the samples, measured on the 70-cc detector, were run using a 1-cc planar Ge(Li) detector having 600-eV resolution at 122 keV. These spectra were also acquired at 3000 counts/sec and with a cadmium absorber in place.

FIGURE 1 - Typical spectrum of weapons-grade plutonium material.

FIGURE 2 - Expanded portion of spectrum of NBS-SRM 948.

²³⁸Pu Isotopics The 153-keV peak is the only isolated peak in this energy region available for the plutonium-238 isotopic determination. The ratio of the 153-keV peak from plutonium-238 to the 144-keV peak from plutonium-239 has given results of the order of 10-15% for 94% plutonium-239. However, in analyzing the spectra of other types of materials with lower concentration of plutonium-239, this peak-pair ratio yielded poorer results; see Table 1. Consequently, the 148-keV peak from plutonium-241 decay was chosen as a suitable peak to use in conjunction with the 153-keV peak. The following formula is used:

$$\frac{{}^{238}\text{Pu}}{{}^{239}\text{Pu}} = 1.185 \left(\frac{I_{153}}{I_{148}} \right) R({}^{241}\text{Pu}/{}^{239}\text{Pu})$$

where I_{153} and I_{148} are the peak areas of the 153-keV and 148-keV peaks, respectively, and $R({}^{241}\text{Pu}/{}^{239}\text{Pu})$ is the ratio of plutonium-241 relative to plutonium-239 in parts per million as determined from the 208-keV/203-keV peak ratio. The results of using this ratio in the analysis of the NBS-Standard Reference Materials 946, 947 and 948 are shown in Column 6 of Table 1.

Problems still exist in the extraction of the 153-keV peak areas for low plutonium-238 concentrations. Peak areas determined by the computer codes GAUSS V and GRPANL for this peak can vary by as much as 15% due to poor signal-to-noise ratios for background selections which are apparently equally valid. This is consistent with the reproducibility predictions in Gunnink's simulation studies for this peak grouping.⁵

²⁴⁰Pu Isotopics The energy region at 160 keV has been investigated to yield the plutonium-240 isotopics. The spectra were analyzed using GRPANL on the following gamma-ray peaks:

Energy (keV)	Isotope
160.00	²⁴¹ Pu
160.35	²⁴⁰ Pu
161.45	²³⁹ Pu
161.60	²⁴¹ Am
164.60	²⁴¹ Pu - ²³⁷ U, ²⁴¹ Am

The ²⁴⁰Pu/²⁴¹Pu ratio is calculated from the areas of the 160.35 and 164.60-keV peaks. The ²⁴⁰Pu/²³⁹Pu ratio is obtained in conjunction with the ²⁴¹Pu/²³⁹Pu ratio obtained from the 208-keV/203-keV peaks as shown in the following equation:

$$\frac{{}^{240}\text{Pu}}{{}^{239}\text{Pu}} = 50.7 \left(\frac{I_{160}}{I_{164}} \right) R({}^{241}\text{Pu}/{}^{239}\text{Pu})$$

The results from this analysis are listed in Table 2 for various samples and two detector systems. The ratios listed are the average ratio of the gamma-ray to mass spectroscopic values obtained for each category of material. The data obtained on the 1-cc Ge(Li) detector with its better resolution yield values in closer agreement with the mass spectroscopic values. This energy region will be investigated further using this spectroscopy system.

²⁴¹Pu Isotopics Two peak pairs (i.e., 148 keV/144 keV and 208 keV/203 keV) have been used extensively in our work. For plutonium-241 ratio determinations for weapons-grade plutonium material, the agreement between these two calculations is usually within 3%. This agreement in fact was used to verify the correctness of the 144-keV peak area for the plutonium-238 calculations. For the Inventory Verification Program, the weighted average of these two calculations has been adopted for the reported gamma-ray-determined plutonium-241 isotopics.

Investigations of lower-fissile material show that the 148-keV/144-keV peak ratios yield poorer results. Table 1 shows the results of these studies on NBS material. The 208-keV/203-keV peak-pair alone is used for plutonium-241 determination at plutonium-239 concentrations less than 90%.

²⁴¹Am Isotopics The americium-241 isotopic ratios are routinely determined for the Metals Exchange and Inventory Verification Programs using the 662-keV/659-keV peaks.¹ Our experience in determining these values has yielded results which are 0.97 ± 0.04 of the radiocounting values obtained in the Metal Exchange Program. Sufficient radiocounting data are not available to make a similar comparison for the Inventory Verification Program. However, the comparison of the gamma-ray-determined isotopics on the bulk sample versus the aliquot using these peak areas gives a ratio of 1.02 ± 0.07 for Inventory Verification samples.

Three new areas have been investigated for use in the determination of the americium-241 isotopics. The first involves using the ratio of the 125-keV peak from americium-241 and the 129-keV peak from plutonium-239. The formula used in the calculation is:

$$\frac{{}^{241}\text{Am}}{{}^{239}\text{Pu}} = 27875 \left(\frac{I_{125}}{I_{129}} \right) - 540$$

Table 1

COMPARISON OF RESULTS FOR OBTAINING PLUTONIUM-238 AND
PLUTONIUM-241 RATIOS USING DIFFERENT PEAK PAIRS

Sample	Detector	Number of Runs	Average $^{238}\text{Pu}/^{239}\text{Pu}$ (in ppm)			Average $^{241}\text{Pu}/^{239}\text{Pu}$ (in ppm)		
			NBS Value	153/144 Peaks	153/148 Peaks	NBS Value	148/144 Peaks	208/203 Peaks
NBS-948 (92% ^{239}Pu)	70 cc	10	118 $\pm$ 11	144 $\pm$ 6 ^a	144 $\pm$ 7 ^a	4670 $\pm$ 10	4690 $\pm$ 84 ^a	4720 $\pm$ 102 ^a
	1 cc	9	118 $\pm$ 11	127 $\pm$ 25	124 $\pm$ 25	4670 $\pm$ 10	4657 $\pm$ 300	4529 $\pm$ 52
NBS-947 (76% ^{239}Pu)	70 cc	7	3850 $\pm$ 80	2810 $\pm$ 76	4010 $\pm$ 60	54580 $\pm$ 70	39030 $\pm$ 1310	55830 $\pm$ 310
NBS-946 (83% ^{239}Pu)	70 cc	10	2925 $\pm$ 83	2129 $\pm$ 130	2935 $\pm$ 170	43690 $\pm$ 55	32030 $\pm$ 2210	44100 $\pm$ 1610

^aErrors are standard deviations of the mean value.

Table 2

AVERAGE RATIOS OF GAMMA-RAY TO MASS SPECTROSCOPY VALUES FOR PLUTONIUM-240 CONCENTRATIONS
(GAMMA-RAY RESULTS ARE OBTAINED USING THE 160-keV REGION)

Sample	Number of Spectra	Detector	Average Ratio Gamma-Ray/MS
Metal Exchange	25	70 cc	0.80 $\pm$ 0.15
Metal Exchange	8	1 cc	0.94 $\pm$ 0.04
Verification Analytical Aliquots	20	70 cc	0.70 $\pm$ 0.13
Verification Analytical Aliquots	15	1 cc	0.95 $\pm$ 0.07

where the constant term arises from the correction to the intensity of the 125-keV peak due to contributions to this peak from the decay of plutonium-239. Results of these calculations using spectra acquired in the Metal Exchange Program yield results which are only 61% of the value obtained from the 662-keV/659-keV peak-pair calculation. The difficulty here is in the extraction of the 125-keV peak area. The background has a steep slope, and interferences from the complex x-ray structure at 120 keV give a poor signal-to-background ratio at 125 keV (see Figure 1). This region will be investigated further using a high-resolution detector and higher gain.

The second area of investigation is the 335-keV peak grouping suggested by Gunnink.⁴ The peaks investigated were:

Energy (keV)	Isotope
332.30	$^{241}\text{Pu} - ^{237}\text{U}, ^{241}\text{Am}$
332.80	^{239}Pu
335.35	$^{241}\text{Pu} - ^{237}\text{U}, ^{241}\text{Am}$
336.06	^{239}Pu

The group was analyzed using GRPANL, allowing the energies of all four peaks to vary in obtaining the fit. The $^{241}\text{Pu}/^{241}\text{Am}$ ratio is calculated from the area of the 332.3 and 335.35-keV peaks and the $^{241}\text{Am}/^{239}\text{Pu}$ ratio obtained using the $^{241}\text{Pu}/^{239}\text{Pu}$ ratio. The formula used is:

$$\frac{^{241}\text{Pu}}{^{241}\text{Am}} = \frac{0.6353 - 2.060 \cdot (I_{332.3}/I_{335.35})}{0.3686 \cdot (I_{332.3}/I_{335.35}) - 3.818}$$

Only the 25 metal exchange samples have been analyzed by this method. The average ratio of this value to the value obtained from the 660-keV peak-pair is 0.88 ± 0.24 .

The third method for determining americium-241 is limited to samples containing more than 1000 ppm. At these concentrations, the 169-keV peak appears above the background and is used in conjunction with the 171-keV peak from plutonium-239. Using the formula

$$\frac{^{241}\text{Am}}{^{239}\text{Pu}} = 11525 \left(\frac{I_{169}}{I_{171}} \right)$$

the average calculated ratio for eight verification samples was 1.09 ± 0.09 of the value obtained with the 660-keV peaks. The investigation of this method will be extended and may become more useful with the analysis of lower-fissile fuel where plutonium-241 and americium-241 are present in higher concentrations.

ATTENUATION AND EFFICIENCY CORRECTIONS

Gamma-ray isotopics are calculated from formulas of the form

$$\frac{N_1}{N_2} = \frac{I_1 \lambda_2 B_2 \epsilon_2 \alpha_2}{I_2 \lambda_1 B_1 \epsilon_1 \alpha_1}$$

where N is the number of atoms of the isotope present in the sample, I is the intensity of the gamma-ray detected in the spectrum, λ is the decay constant of the isotope, B is the branching ratio, ϵ is the detector efficiency, and α is the absorption and attenuation factor for the gamma ray of interest.

The analysis used in determining plutonium and americium isotopics is based on the assumption that gamma-ray attenuation and detector efficiency can be made negligible by choosing gamma-ray peak pairs whose energies are less than 10 keV apart. Over this energy range both effects are slowly and smoothly varying functions of energy. Thus, over the 10-keV energy interval, the ratio $(\epsilon_2 \alpha_2 / \epsilon_1 \alpha_1)$ is assumed to be equal to one. The formula then reduces to

$$\frac{N_1}{N_2} = \frac{I_1 \lambda_2 B_2}{I_2 \lambda_1 B_1}$$

During this report period, the shape of the efficiency-attenuation curve has been determined by analyzing the 129, 144, 171, 189, and 203-keV peaks from the decay of plutonium-239. The curve obtained by dividing the areas of these peaks by their corresponding branching ratio yields a curve proportional to the product of the attenuation of the gamma-rays and the efficiency of the detector for the sample. Correction factors were taken from this curve and applied to the areas of the peaks used for the isotopic measurements. Figure 3 shows the curve for the two detector systems used to obtain the spectrum of a Metal Exchange sample. The variation of the shape of the curve, in this case, is due to the detector. Variations also arise with changes in sample size, composition, packaging materials and/or absorbers. Data for these studies have been acquired during the December 1974 and February 1975 inventory verification series and during recent metal exchange measurements. These data cover a wide range of matrix categories and sample weights. Preliminary results show that such corrections are more important for metal and oxide samples than for scrap categories. To date, final results are available for one set of spectra on small samples and are discussed in the subsequent section.

FIGURE 3 - Gamma spectra for two detector systems.

These corrections cannot be applied to earlier data because insufficient peak information was extracted at the time these earlier spectra were being analyzed.

Results

SMALL PLUTONIUM METAL AND OXIDE SAMPLES

The spectra from 34 nominal 1-g plutonium metal, plutonium oxide and mixed plutonium oxide samples were analyzed using the program GAUSS V. The purpose of this exercise was to investigate the bias in the $^{238}\text{Pu}/^{239}\text{Pu}$ atom ratio and the correlation of the bias with increasing concentration of plutonium-238 as reported previously.¹ The range of plutonium-238 concentration was 50 to 350 ppm as determined by radiocounting slides prepared from the samples. The least-squares fit to the line drawn through these data points plotted as the ratio of the plutonium-238 concentration determined by gamma-ray spectroscopy (NDA)/radiocounting (DA) versus plutonium-238 concentration determined by radiocounting is summarized in Table 3. The parameters a and b in Table 3 are defined in the equation:

$$\frac{^{238}\text{Pu}}{^{239}\text{Pu}}(\text{NDA}) / \frac{^{238}\text{Pu}}{^{239}\text{Pu}}(\text{DA}) = a + b[^{238}\text{Pu}(\text{DA})]$$

The reported results were obtained from the peak areas obtained by simple peak area summation over the interval of the peak; GAUSS V (uncorrected) results were obtained from the fitted peaks obtained from GAUSS V; GAUSS V (corrected) results were obtained by applying attenuation and efficiency corrections to the peaks obtained from the GAUSS V fitting. Because of the poor statistics involved in obtaining the plutonium-238 isotopic ratios for those samples with plutonium-238 concentrations less than 100 ppm, the correlation function was repeated for each set of data excluding those points to give the six sets of data in the table. Zero bias and correlation would yield a value of $a = 1$ and $b = 0$.

For all methods of analysis the slope of the correlation line disappears when the data points below 100 ppm plutonium-238 are ignored. The best precision for the line is obtained in using GAUSS V areas corrected to include attenuation and detector efficiency.

The attenuation and efficiency corrections, determined by analyzing the 129, 144, 171, 179, 189, and 203-keV peaks from the decay of plutonium-239, were applied to the areas of the 144-keV and 153-keV peaks to give the corrected values of the

Table 3

CORRELATION CONSTANTS FOR COMPARISON OF ^{238}Pu (NDA) to ^{238}Pu (DA)
ON ONE-GRAM SAMPLES WITH PLUTONIUM-238 RANGE FROM 50 TO 350 PPM

Method of Analysis	Number of Data Points	Correlation Constants	
		a	b
Reported Results ^a	34	1.1366 ± 0.0624	-0.0010 ± 0.0003
Reported Results (Points >100 ppm ^{238}Pu)	23	0.9185 ± 0.0679	-0.0002 ± 0.0003
GAUSS V	34	1.3922 ± 0.0691	-0.0012 ± 0.0003
GAUSS V (Points >100 ppm ^{238}Pu)	23	1.1509 ± 0.0657	-0.0003 ± 0.0003
GAUSS V (Corrected)	34	1.1376 ± 0.0583	-0.0009 ± 0.0003
GAUSS V (Corrected) (Points >100 ppm ^{238}Pu)	23	0.9727 ± 0.0523	-0.0002 ± 0.0002

^aFrom Reference 1.

plutonium-238 ratio used in the last two entries of Table 3.

METAL EXCHANGE SAMPLES

Thirty metal exchange samples have been analyzed during the past 2 yr on the 70-cc Ge(Li) spectrometer. The comparison of the NDA results achieved by gamma-ray spectroscopy as obtained from GAUSS V are compared to the DA results:

Isotopic Ratio	Average Ratio NDA/DA	
	GAUSS V	GRPANL
$^{238}\text{Pu}/^{239}\text{Pu}$	1.09 ± 0.15	0.96 ± 0.12
$^{240}\text{Pu}/^{239}\text{Pu}$	0.94 ± 0.05 ^a	0.80 ± 0.15 ^b
$^{241}\text{Pu}/^{239}\text{Pu}$	1.04 ± 0.05	1.02 ± 0.04
$^{241}\text{Am}/^{239}\text{Pu}$	0.97 ± 0.04	-

^aGAUSS V analysis of peaks at 640 keV.

^bGRPANL analysis of peaks at 160 keV.

In addition, eight samples were also analyzed using a 1-cc Ge(Li) spectrometer, a 0.030-in. (0.076-cm) cadmium absorber and amplifier gain of 0.12 keV per channel. Analysis of the spectra was performed using GRPANL. The plutonium-240 concentration was calculated using the 160-keV region. The results are:

Isotopic Ratio	Average Ratio NDA/DA GRPANL
$^{238}\text{Pu}/^{239}\text{Pu}$	1.00 ± 0.24
$^{240}\text{Pu}/^{239}\text{Pu}$	0.94 ± 0.04
$^{241}\text{Pu}/^{239}\text{Pu}$	1.00 ± 0.02

The range of plutonium-238 concentrations is 110 to 180 ppm. No correlation in bias as a function of concentration is evident over this concentration range.

NATIONAL BUREAU OF STANDARDS - STANDARD REFERENCE MATERIAL

Seven to ten spectra were acquired using the 70-cc Ge(Li) spectrometer for each of the NBS Standard Reference Materials 946, 947, and 948. In addition, nine spectra were acquired for NBS-SRM 948 using the 1-cc Ge(Li) detector. For these measurements a cadmium absorber was used and the count rate was maintained at approximately 3000 counts/sec. The data were reduced using the program GAUSS V. The values in Table 4 for the isotopic content are the averages of the values obtained from each spectrum. The errors are the standard deviation of the mean. The plutonium-238 values are in good agreement with the certified value except for NBS-SRM 948. Here a large difference arising from the poor signal-

Table 4

COMPARISONS OF ISOTOPIC RATIOS DETERMINED BY GAMMA-RAY SPECTROSCOPY
TO THE CERTIFIED VALUES FOR NBS - STANDARD REFERENCE MATERIALS

Sample	$^{238}\text{Pu}/^{239}\text{Pu}$ (in ppm)			$^{240}\text{Pu}/^{239}\text{Pu}$ (in ppm)			$^{241}\text{Pu}/^{239}\text{Pu}$ (in ppm)		
	γ -Ray	NBS Value	Ratio	γ -Ray	NBS Value	Ratio	γ -Ray	NBS Value	Ratio
NBS-946	2935 $\pm$ 170	2925 $\pm$ 83	1.00				44100 $\pm$ 1640	43690 $\pm$ 60	1.01
NBS-947	4010 $\pm$ 60	3850 $\pm$ 80	1.04	198260 $\pm$ 37660	241560 $\pm$ 300	0.83	55830 $\pm$ 310	54580 $\pm$ 70	1.02
NBS-948 [75 cc Ge(Li)]	144 $\pm$ 7	118 $\pm$ 11	1.22	96250 $\pm$ 7880	86410 $\pm$ 110	1.11	4720 $\pm$ 102	4670 $\pm$ 10	1.01
NBS-948 [1 cc Ge(Li)]	127 $\pm$ 25	118 $\pm$ 11	1.07				4529 $\pm$ 52	4670 $\pm$ 10	0.97

to-background noise associated with the 153-keV peak exists. Somewhat better agreement is obtained from the 1-cc Ge(Li) detector data. This is due to better resolution obtained with the smaller detector and a flatter slope in the background under the 153-keV peak. The plutonium-241 ratio is within 3% of the certified value provided the 208-keV/203-keV peak pairs are used for the calculations. The $^{240}\text{Pu}/^{239}\text{Pu}$ determinations are poor because of the poor statistics in the acquisition of the data. Under these experimental conditions, clearly greater than 8-hr counting times for the 0.25-g samples are necessary to obtain adequate statistics in the 640-keV region for good plutonium-240 determinations.

Additional work is presently being performed to acquire spectra with significant data in the 640-keV region for plutonium-240 determinations. In addition, data will be acquired on the 1-cc detector for each reference material and these spectra will be reduced using GRPANL.

ALO INVENTORY VERIFICATION SAMPLES

The isotopic ratios necessary for calorimetric assay have been determined for 50 bulk samples and 28 aliquot samples received under the Inventory Verification Program. These analytical samples are taken from the bulk samples at the time of packaging for shipment to Mound to aid in the determination of the isotopic ratios and to establish further reliability upon the NDA techniques by enabling a comparison between destructive analysis

and gamma spectroscopy. Table 5 is a summary of the comparison of the non-destructive gamma-ray assay and the chemical assay of the aliquot samples and the comparison of the aliquot samples to their bulk sample parents. The spectra were acquired with the 70-cc Ge(Li) detector with a 0.030-in. (0.076-cm) cadmium absorber. The computer program GAUSS V was used to extract the peak areas. The peak areas used in the isotopic determinations are: 153 keV/144 keV for the $^{238}\text{Pu}/^{239}\text{Pu}$ ratio; 642 keV/640 keV for the $^{240}\text{Pu}/^{239}\text{Pu}$ ratio; 208 keV/203 keV and 148 keV/144 keV for the $^{241}\text{Pu}/^{239}\text{Pu}$ ratio; and 662 keV/659 keV for the $^{241}\text{Am}/^{239}\text{Pu}$ ratio.

The verification samples have been analyzed in several additional experiments designed to improve the accuracy and/or the reproducibility of the isotopic ratios. These tests can be classified under two major headings: fitting techniques and sample collimation.

Fitting Techniques

The fitting program GAUSS V only accepts linear backgrounds. Over small energy regions this assumption of a linear background should be a good approximation. The uncertainty in the background, however, can be a major contributor to the uncertainty in the peak area. For these reasons the general purpose gamma-ray peak-fitting programs of GRPANL have been adapted to run on a PDP-8L computer.

The 20 samples and 20 aliquots from the February 1975 verification have been

Table 5

COMPARISON OF GAMMA-RAY ISOTOPIC RATIOS TO CHEMICAL ISOTOPIC RATIOS
(VALUES STATED ARE THE AVERAGE RATIOS AND THEIR
STANDARD DEVIATION FOR THE NUMBER OF SAMPLES ANALYZED)

	Plutonium Inventory Series	
	December 1974 (8 samples)	February 1975 (20 samples)
$^{238}\text{Pu}/^{239}\text{Pu}$		
Gamma Aliquot/MS	1.05 ± 0.09	1.22 ± 0.16
Gamma Aliquot/Radiocounting	1.00 ± 0.05	1.11 ± 0.14
Gamma Can/Gamma Aliquot	1.07 ± 0.16	1.43 ± 0.27
$^{240}\text{Pu}/^{239}\text{Pu}$		
Gamma Aliquot/MS	0.90 ± 0.06	0.94 ± 0.11
Gamma Can/Gamma Aliquot	0.92 ± 0.09	1.03 ± 0.11
$^{241}\text{Pu}/^{239}\text{Pu}$		
Gamma Aliquot/MS	0.97 ± 0.02	1.01 ± 0.02
Gamma Can/Gamma Aliquot	0.98 ± 0.04	1.01 ± 0.01
$^{241}\text{Am}/^{239}\text{Pu}$		
Gamma Can/Gamma Aliquot	1.04 ± 0.07	1.01 ± 0.06

analyzed using GRPANL programs. Several additional peak pair sets have been used for the isotopic ratios. They are: 153 keV/144 keV and 153 keV/164 keV for the $^{238}\text{Pu}/^{239}\text{Pu}$ ratio; 160 keV/161 keV and 160 keV/164 keV for the $^{240}\text{Pu}/^{239}\text{Pu}$ ratio; and 208 keV/203 keV and 148 keV/144 keV for the $^{241}\text{Pu}/^{239}\text{Pu}$ ratio.

Table 6 shows a summary of the comparison of isotopic ratios calculated using the peak areas from GRPANL and chemical assay. The comparison of the GAUSS V results to chemical assay is also presented.

SAMPLE COLLIMATION

Six of the February 1975 samples were used in an experiment in which the detector views only a 0.25-in.-wide segment of the sample as it is rotated past the detector. Three spectra were obtained

with each sample and two detectors were used: the 1-cc Ge(Li) and the 70-cc Ge(Li). Table 7 shows the peak-to-background ratios for one of the samples.

The data from the 70-cc Ge(Li) detector were analyzed using GAUSS V and standard techniques. The isotopic ratios determined by gamma-ray assay, with and without the lead collimator, were compared to the ratios determined by chemical assay. For the $^{238}\text{Pu}/^{239}\text{Pu}$ ratio the collimated values were approximately 20% closer to the chemical assay values than were the uncollimated values. For the other isotopic ratios a small improvement is noted in the standard deviation of the collimated results. Since reduction in background can improve reproducibility and allow shorter counting times, other methods for reducing the background are being studied.

Table 6

AVERAGE OF ISOTOPIC RATIOS USING GAUSS V AND GRPANL PROGRAM (FEBRUARY 1975 VERIFICATION)

Isotopic Ratio	Peak Pair	Aliquot (NDA)/(DA)		Can/Aliquot	
		GAUSS V	GRPANL	GAUSS V	GRPANL
238/239	153/144	1.11 ± 0.14	0.94 ± 0.08	1.43 ± 0.27	1.18 ± 0.17
	153/164	-	0.99 ± 0.08	-	1.18 ± 0.18
240/239	160/161	-	0.84 ± 0.18	-	0.86 ± 0.22
	160/164	-	0.70 ± 0.13	-	0.71 ± 0.09
	642/640	0.94 ± 0.11	-	0.97 ± 0.11	-
241/239	- ^a	1.01 ± 0.02	1.02 ± 0.03	1.01 ± 0.01	1.01 ± 0.03

^aThe 241/239 ratio was obtained using the weighted average of the 208/203 and the 148/144 peak pairs.

Table 7

PEAK-TO-BACKGROUND RATIOS

Energy (keV)	1-cc Ge(Li)		70-cc Ge(Li)	
	No Shield	Shield	No Shield	Shield
129	10:1	15:1	9:1	11:1
148	3:1	4:1	2.8:1	3.4:1
208	14:1	16:1	18:1	19:1

Systems Development

AUTOMATED PLUTONIUM ASSAY SYSTEM

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INTRODUCTION

The Automated Plutonium Assay System (APAS) is being developed as an accurate in-line assay system for mixed oxide fuels. A process control computer will direct and control the movement of the samples from the storage area to measurement stations. The data acquired by the computer will be immediately processed and will help provide material control and safeguards accountability. An isometric view of a basic calorimetric assay system installed in a glovebox line is shown in Figure 4.

AUTOMATION

Three-Dimensional (X,Y,Z) Transport

The fabrication of a three-dimensional (X,Y,Z) transport and positioning subsystem (see Figure 5) has been completed and the system installed in a test frame to simulate the glovebox line. It has been designed to move samples between the storage and measurement stations of the APAS.

Installation and synchronization of drive motors and gearing are in progress to insure minimum backlash in the positioning

FIGURE 4 - Basic calorimetric assay system.

FIGURE 5 - Three-dimensional (X,Y,Z) transport and position subsystem.

mechanism. The calculated accuracy of positioning in the three axes of motion is as follows:

X axis: ± 0.004 in. (± 0.010 cm)
Y axis: ± 0.001 in. (± 0.002 cm)
Z axis: ± 0.001 in. (± 0.002 cm)

A weight reduction of approximately 250 lb (113 kg) has been realized by using a stainless steel post construction instead of a solid backed "U" frame for the support rails. The post design provides adequate support and strength for the positioning trolley and will exert less strain on the assay gloveboxes.

Fabrication of a computer-controlled three-axis stepping-motor drive system has been completed, and the system is being installed in the X,Y,Z mechanism. A device for adapting fiber-optic control to the Z axis to detect the presence of a sample has been designed and is being fabricated.

A flat 20-conductor electrical cable layout system is being developed to supply electrical power and control circuits to the X,Y,Z positioning trolley. The cable will be released and retracted by constant-tension devices on each track of the trolley support rails. Preliminary tests indicate that a force of 10 lb (4.5 kg) is required on each flat cable to provide a smooth retraction during movement of the trolley.

Transport System

A preliminary design study for an external transport system to move samples between unit-process and the assay system was conducted. The preliminary design favors a computer-controlled mechanized conveyor system within a closed duct, thereby insuring the integrity of the RETIMAC concept.

Computer

Delivery has been made of the process-control computer for the assay system (a PDP-11/40*). The installation and hardware interfacing is in progress. RSX-11M* has been purchased as the operating system. Preliminary flow diagrams for material movement through the assay system have been made.

Container Identification System

The requirements for an automated unit identification system are being defined.

*Digital Equipment Corporation trademark.

Consideration is being given to both fully automatic and semi-automatic reading stations. Several companies which produce commercial identification systems have been contacted. Their marking systems are being evaluated for tamper resistance and compatibility with packaging requirements for the assay system and with the overall RETIMAC concept.

One set of permanent and peelable adhesive marking labels has been obtained. Tests on the "permanent" labels affixed to metallic surfaces show that these labels can be removed by immersing in Freon TF. If the label is allowed to dry, it can be replaced such that its prior removal cannot be easily detected. Possible modification of this label and evaluation of other marking techniques is continuing.

Pickup Device

Two dual-state pickup devices are being evaluated. One is commercially available and the other has been built at Mound. An investigation is in progress to determine the compatibility of these devices with calorimeters, the X,Y,Z system, the transport system, and with the requirements of a fuel processing line.

Reliability testing of one of these pickup devices for the SNM containers is in progress in an effort to provide the most dependable unit possible for this function. This reliability test consists of constant and repeated cycling of the pickup/release mechanism with a simulated container to establish a realistic failure rate.

CALORIMETRY

Process Calorimeters

The process calorimeters have been designed to operate in a fully automated in-line assay system. A cross-sectional view of the safeguards calorimeter is shown in Figure 6. The sample area is large enough to hold a 1-kg mixed oxide sample in a 211/209 x 6-7/16 in. can. This calorimeter uses a twin resistance bridge with the reference area below the sample area. This design allows twice as many calorimeters in the glovebox line as the conventional side-by-side design.

The air gap of the calorimeter (see Figure 6) is the principal thermal resistance of the calorimeter. The greater the thermal resistance the higher the sensitivity and the more time required for the calorimeter to reach equilibrium. This gap can be filled with any gas or solid to provide

FIGURE 6 - Cross section of Safeguards calorimeter.

the proper combination of sensitivity and equilibrium time.

When solids are used it is necessary to assure a uniform filling to avoid gradients in the thermal resistance of the gap. Tests have been performed on various mixes of silicone greases, oils, and banding sand to test their mechanical properties when they fill the gap. Banding sand, which has a thermal conductivity nine times that of air, has been selected for the original filling of the calorimeters.

Two process calorimeters have been fabricated (see Figure 7). Before the air gaps of these calorimeters were filled some limited tests were performed to measure the basic sensitivity and half-lives. Since sand has approximately the same thermal characteristics as mixed oxide, the sample used in these tests consisted of a can containing a plutonium-238 heat source packed in sand. The calorimeter sensitivity (output per unit power input) is approximately 80,000 $\mu\text{V/W}$. The reproducibility of the bridge potential is 0.02% while the stability of an individual reading is less than 0.02%.

When installed, the calorimeters will be immersed in two separate water baths. Electrical controllers will be associated with the bath to control the temperature to $\pm 0.001^\circ\text{C}$. Water baths will also be installed under the storage region to pre-equilibrate samples, thereby reducing the calorimeter equilibrium time. In a further effort to reduce the equilibrium time, the calorimeters will be servo controlled; that is, the calorimeter will be maintained at a constant temperature by applying electrical power to the servo heater. The measured sample power is then equal to the reduction in the electrical power applied to the servo heater to maintain the set point temperature.

Analytical Calorimeter

To provide a more accurate watts-per-gram conversion than can be obtained from isotopic determinations, the feasibility of an analytical calorimeter is being studied. The calorimeter would measure the power of small quantities of plutonium which would then be dissolved and chemically assayed. Additional uses of the instrument are to empirically determine the increase of power versus time due to ingrowth of

FIGURE 7 - Process calorimeter.

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americium-241 and to determine the plutonium content of mixed oxides by calorimetry and weighing.

Ideally, the sample size for the specific power determination should not be over 1 g to be consistent with chemical assay techniques. It would also be preferable to have calorimetric assay 10 times more sensitive than chemical assay. As a result, the uncertainty of the measurement would depend primarily on the uncertainty of chemical assay. Assuming 5 mW/g and precisions of $\pm 0.1\%$ for chemical assay, this would mean calorimeter precisions of $\pm 0.5 \mu\text{W}$. To date, this level of precision has only been achieved on a research basis. Thus, some compromises are necessary.

At this point two basic alternatives exist. One is to develop a small-volume (~1 cc) calorimeter for specific power and power increase determinations, then to use existing calorimeter designs to determine plutonium concentration in mixed oxide fuels. The second choice is to attempt to develop a single calorimeter to do all three jobs. In either case a new calorimeter will need to be developed. At this point the most attractive alternative is to have two-calorimeters since the 1-cc calorimeter would be significantly faster than the larger volume unit. This would also provide more flexibility since plutonium concentrations ranging from 0.2 to 20% are possible in future nuclear fuels.

The small analytical calorimeter would have to be placed in the analytical lab to provide the type of environmental conditions necessary for the accuracies and precisions required.

Cans

The design of a reusable stainless steel calorimeter can is being investigated. This study is closely linked with the design considerations of the calorimeter baffles and the sample pickup device. The criteria for the cans are that they have a tamper-proof seal, are reusable, are approximately 6 in. (15 cm) high by 2 in. (5 cm) in diameter, and are compatible with an automated identification system, the assay system's X,Y,Z mover, and the transport system.

Calorimeter Equilibrium Time

The process calorimeter system is being designed and built with the idea of obtaining equilibrium in 1 hr. Sample pre-equilibration and isothermal servo control of the calorimeter are two physical

techniques which can be utilized to achieve this goal. These techniques are being incorporated into the system. However, end point prediction studies are being conducted as an alternative approach. The prediction studies have shown that a general expression of the equilibrium curve cannot be derived for all calorimeters or all samples since the calorimeter, sample matrix, sample heat capacity, fuel density, and sample container are all critical parameters in determining the algorithm.

GAMMA-RAY ISOTOPICS

Hardware

The multichannel analyzer and its interface to the assay computer have been received. An intrinsic germanium detector is being evaluated to see if it is possible to determine all the isotopic ratios reliably from gamma-ray peak pairs less than 350 keV.

Software

Several general-purpose gamma-ray analysis programs written in FORTRAN II have been added to the PDP-8 computer libraries and have been modified to run without operator intervention. These programs have also been linked to another FORTRAN-II program which calculates isotopic ratios using the peak areas. These studies will help in the design of the analysis programs for the assay system. Several categories of plutonium have been measured. The best set of peak pairs is dependent on the category; therefore the programs will have to be modified to compensate for the fuel characteristics.

SPONTANEOUS FISSION ASSAY SYSTEM (SFAS)

In order to facilitate the addition of a spontaneous fission assay system (SFAS) to the original assay system, LASL has been contacted and has agreed to help define the requirements of an SFAS. This includes supplying design criteria and operating procedures. Whenever possible, all hardware will be obtained commercially. It is hoped that some of the software being developed by LASL for their coincidence counters can be adapted with minimal change to run on the assay computer.

ENGINEERING

Drawings of conceptual designs for retrofitting the assay system at HEDL have been prepared.

The original three-glovebox system has been received. The gloveboxes have been

leak tested with a helium mass spectrometer and meet Mound Laboratory specification 1-14425. They have been assembled and are being prepared for installation of the X,Y,Z system. Figure 8 is a photograph of the three-glovebox system.

A fourth glovebox will be necessary in the expanded system to house a

spontaneous fission assay system and will be used to interface the X,Y,Z mover of the assay system to the transporter. Preliminary drawings for this box have been made and are being reviewed. An isometric view of a four-glovebox assay system containing a neutron well counter is shown in Figure 9.

FIGURE 8 - Three-glovebox system.

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FIGURE 9 - Four-glovebox assay system.

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