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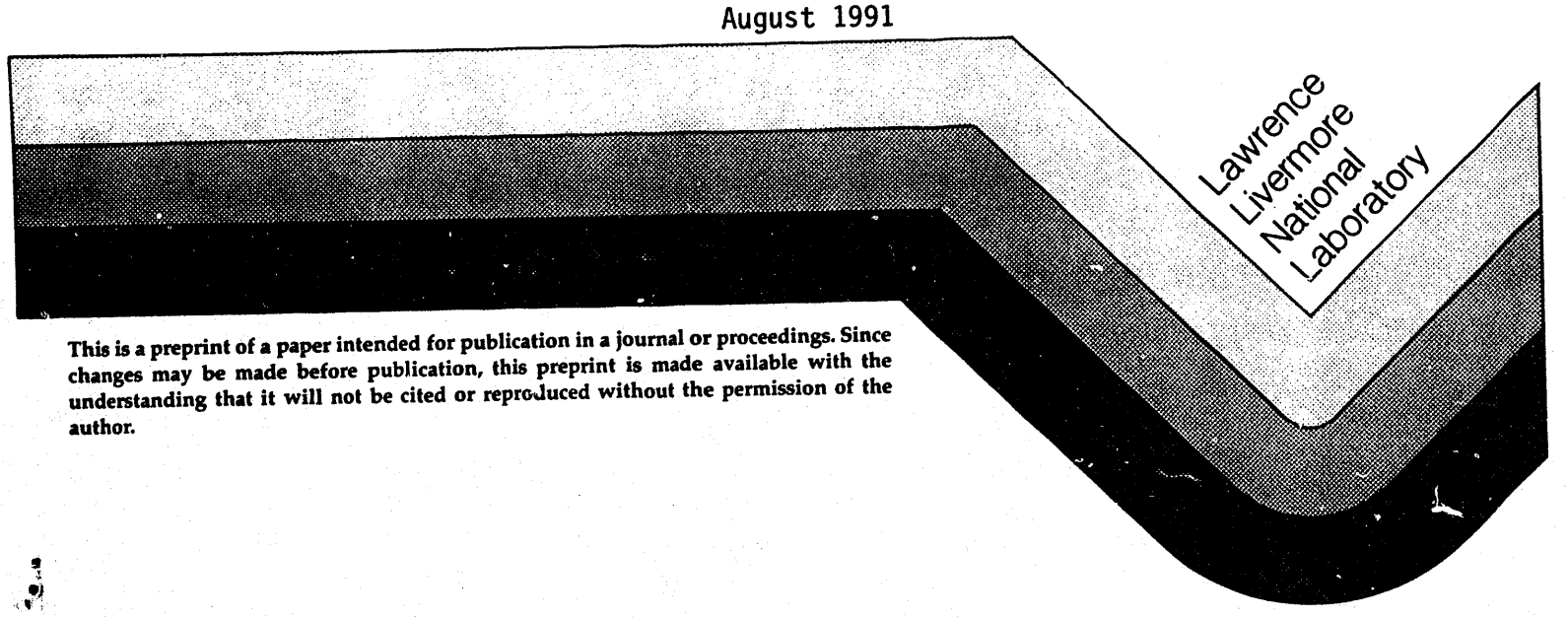

AN ISOTOPIC ANALYSIS SYSTEM FOR PLUTONIUM SAMPLES ENRICHED IN ^{238}Pu*

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ABSTRACT

We have designed and built a gamma-ray spectrometer system that measures the relative plutonium isotopic abundances of plutonium oxide enriched in ^{238}Pu. The first system installed at Westinghouse Savannah River Company was tested and evaluated on plutonium oxide in stainless steel EP60/61 containers. ^{238}Pu enrichments ranged from 20% to 85%. Results show that 200 grams of plutonium oxide in an EP60/61 container can be measured with $\pm$0.3% precision and better than $\pm$1.0% accuracy in the specific power using a counting time of 50 minutes.

INTRODUCTION

Plutonium enriched in ^{238}Pu is used as a source of heat and power (0.57 W/g). Radioisotope thermoelectric generators (RTG) using plutonium enriched in ^{238}Pu are used as sources of power for space-exploration vehicles, satellites, etc. Enriched plutonium materials exist in the DOE complex and at other government agencies, e.g., at the National Aeronautics and Space Administration (NASA). To account properly for this material, it is necessary to determine the amount of fissile isotopes of plutonium, i.e. ^{239}Pu and ^{241}Pu present in the enriched material. Gamma-ray spectrometry can nondestructively determine the relative plutonium abundances of the various isotopes. Although the gamma-ray spectrum is dominated by gamma rays from ^{238}Pu, there are several weak gamma-ray lines that can be used to determine the two fissile isotopes of plutonium and of americium-241.

The MGA code[1], which is used for the isotopic analysis of plutonium materials with normal reactor burnups, cannot be used for the analyses of gamma-ray spectra from these ^{238}Pu materials, because the 100-keV region used by MGA is obscured by intense ^{238}Pu gamma rays. We have developed a new analysis approach for these spectra using twelve selected gamma rays from 152 to 860 keV. This new analysis method uses the GRPANL[2] code to determine gamma-ray peak intensities and the methodology of the MGA code to determine the effects of plutonium self-absorption and the relative plutonium isotopic abundances.

The plutonium oxide is doubly contained in two cylindrically-shaped, stainless-steel containers, referred to as EP60/61 containers. The volume containing the plutonium material is approximately 2.6 cm in diameter by 30.5 cm in length and may contain over 200 grams of oxide. The total wall thickness of the two containers is 1.2 cm. Sample amounts will vary and the location of the material within the container volume is unknown. Therefore, the entire length of the container must be measured from all directions. ^{238}Pu oxide emits significant amounts

of neutrons from (α,n) interactions. Neutron emission intensities of this magnitude impacts instrumentation design from considerations of both personnel radiation safety and probable neutron damage to the gamma-ray detector.

SYSTEM DESIGN

Figure 1 shows a cross-sectional view of the sample enclosure and detector shield we have designed to measure plutonium materials enriched in ^{238}Pu. A 20% efficient, coaxial, high-purity germanium (HPGe) detector views the sample from a distance of about 28 cm through a 7-cm diameter window in the sample-shielding enclosure. The window contains a graded-absorber assembly (0.98 g/cm^2 Pb, 1.50 g/cm^2 Cd, and 0.45 g/cm^2 Cu) to reduce the intensity of the strong 99.86- and 152.68-keV gamma rays from ^{238}Pu; and 5 cms of borated polyethylene is used to reduce the exposure of the detector to neutrons emitted by a sample. Lead shielding, that is 5-cm thick, completely surrounds the detector window inside the enclosure to prevent the detector from "seeing" high-energy gamma-rays that do not pass through

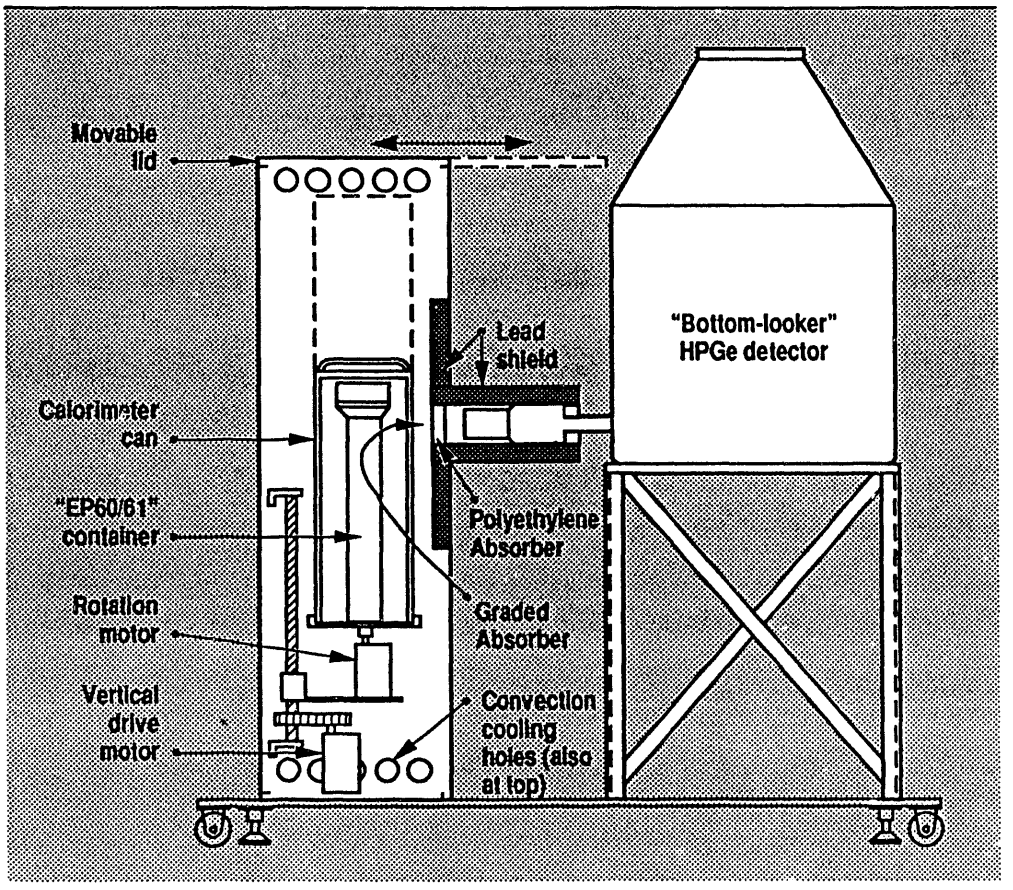

Fig. 1 During each measurement, the flashlight-shaped EP60/61 container seen inside the sample enclosure moves continually up and down while rotating, so that the detector measures the entire sample.

the window. A 5-cm-thick lead barrel shields the detector from sample gamma rays other than those passing through the window and from other samples that may be nearby.

An elevator mechanism inside the enclosure raises and lowers a sample for easy loading and unloading. This mechanism is used during a measurement to vertically scan the entire length of a sample container up and down in front of the HPGe detector. Simultaneously, the container is rotated so that its entire contents are viewed by the detector during a measurement. Sensors are placed on both the vertical- and rotation-drive-motor shafts to insure their continuous operation during a measurement. If either the rotation or scanning motion stops, the measurement stops and an error message is displayed on the operator's terminal. The sample-enclosure lid must be closed before a measurement can begin and is prevented from being opened during a measurement. This is done to prevent the

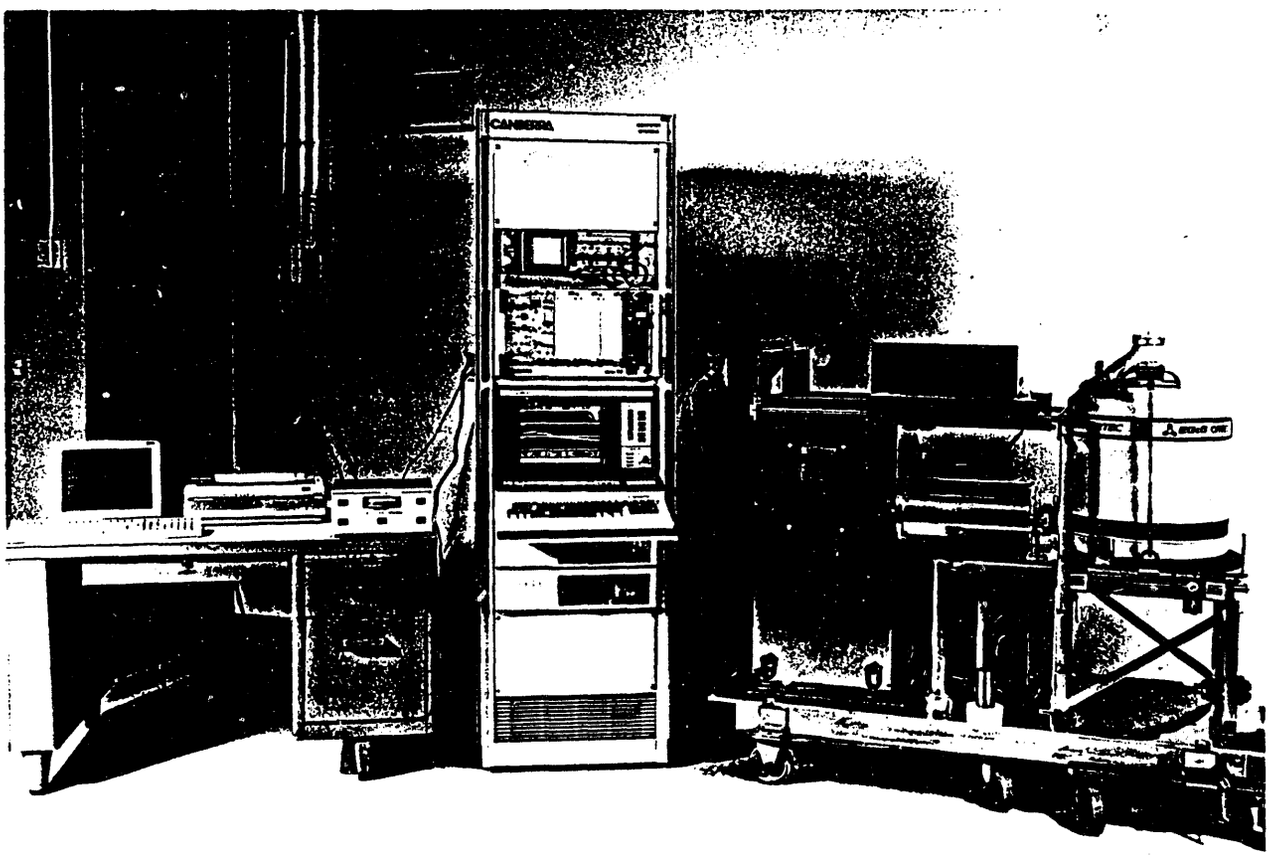

Fig. 2 Photograph of the Westinghouse Hanford Company ^{238}Pu gamma-ray spectrometer system and its supporting electronics.

inadvertent exchange of a sample during a measurement, or corruption of the gamma-ray spectrum being measured by background gamma rays.

The enclosure has substantial shielding to protect personnel from both the gamma radiation and neutrons that are emitted from the ^{238}Pu materials. The sample consists of 5-cm-thick borated polyethylene for neutron shielding plus 1.25 cm each of lead and stainless steel to reduce external exposure from a sample's gamma-ray emissions. Because the samples are thermally hot (they can generate more than 100 W), holes are located at the top and bottom on three sides of the sample-shielding enclosure to allow for convective cooling.

Figure 2 is a photograph of the entire system. The operator's terminal is on the desk on the left along with a printer for output of measurement results. On the right-hand side of the desk is the instrument-control box used by an operator to open the enclosure and elevate the turntable for (un)loading a sample. The control box has a meter that is delimited, to indicate the system's counting deadtime, with three colored regions: yellow, 0-10%, TOO LOW; green, 10-40%, OPTIMUM; and red, 40-100%, TOO HIGH. The detector can be moved closer to, or away from, a sample by up to 10 cm to better optimize its counting.

The detector amplifier is operated with one microsecond Gaussian shaping to optimize gamma-ray pulse throughput, and reduce the probability of pulse pileup. While this slightly worsens energy resolution in the gamma-ray spectrum, energy resolution is not a prerequisite for the spectrum analysis, because most of the gamma-ray intensities are determined by integrating net counts in each channel rather than peak fitting. Pulses from the amplifier are digitized with a 100-Mhz analog-to-digital converter (ADC) and stored in a 4096-channel multichannel analyzer. Once a measurement is completed, the spectral data are automatically transferred from the multichannel analyzer to a Digital Equipment Corporation MicroVAX II computer where they are stored on disk and analyzed for the relative plutonium isotopic abundances.

DATA-ANALYSIS METHODOLOGY

We have selected the twelve gamma rays that are shown in Table 1 to determine the relative plutonium isotopic abundances. The 152.7- and the 766.4-keV gamma-rays are used to measure the amount of ^{238}Pu in the sample. The 208-keV line from the decay of ^{237}U is used to determine the amount of ^{241}Pu; it is assumed that ^{237}U is in equilibrium with ^{241}Pu. The spectral peaks at 375 and 413.7 keV are used to determine the amount of ^{239}Pu. The ^{228}Th gamma rays are used in addition to the plutonium gamma rays to establish the relative detection efficiency.

Table 1. List of gamma rays chosen for the isotopic analysis of materials enriched in ^{238}Pu.

Energy (keV)	Isotope
152.7	^{238}Pu
208.0	^{241}Pu, ^{241}Am
238.6	^{228}Th
375.0	^{239}Pu
376.6	^{241}Am
413.7	^{239}Pu
583.1	^{228}Th
727.4	^{228}Th
742.8	^{238}Pu
766.4	^{238}Pu, ^{228}Th
786.3	^{238}Pu, ^{228}Th
860.5	^{228}Th

The intensities of these twelve selected gamma rays are determined from each measured gamma-ray spectrum by using the computer program GRPANL.[2] The 238-, 375-, 414-, 583-, and 727-keV peak areas are determined by fitting these peaks and other nearby interfering peaks to a Gaussian peak shape with an exponential low-energy tail. All remaining peak areas are determined by simple integration of the net channel counts. After determination of the net peak area, GRPANL corrects them for the absorption effects of the sample container (stainless-steel) and the absorbers used to reduce the intensities of the strong low-energy ^{238}Pu gamma rays.

To determine the relative isotopic abundances the corrected gamma-ray intensities are passed to a separate program that uses an equation that describes the sample self attenuation and relative detection efficiency of the detector. This equation is:

$$A_j = \sum_{k=1,5} (B_{jk} \cdot X_k) \times \frac{1}{\mu_j \cdot PU} (1 - EXP(-\mu_j \cdot PU)) \times eff \cdot (1 + bE_j + cE_j^2),$$

where

A_j = intensity of peak j,

B_{jk} = gamma-ray branching of peak j of component k,

X_k = disintegrations per minute of component k,

μ_j = mass absorption coefficient of plutonium for peak j,

PU = average thickness (g/cm^2) of plutonium in the sample,

eff = estimated detector efficiency for peak j determined from a "standard" efficiency curve,

E_j = energy (keV) of peak j, and

b,c = coefficients in a quadratic function used to account for small deviations in the efficiency from the "standard" value.

This equation is solved by the method of iterative, non-linear, least-squares using the twelve corrected gamma-ray intensities. Its solution gives the plutonium sample thickness and the relative abundances of the plutonium isotopes. The iterative process begins with the disintegration rates (X_k k=1,5) in the sample for the ^{238}Pu, ^{239}Pu, ^{241}Pu, ^{241}Am, ^{228}Th isotopes and the plutonium thickness (PU) in the sample as free variables to be determined by the fit. Once the change in the PU variable during the fitting process is less than 0.1 g/cm^2, the b and c variables that describe small corrections to the relative detector efficiency are freed for the remainder of the fit to the twelve gamma-ray intensities. It should be noted that the b and c coefficients in the fit not only make adjustments for small changes the detector efficiency from "standard" detector efficiency, but also accounts for any small attenuations that may be due to unknown low-Z materials in a sample, which will be manifested as small changes in the efficiency curve.

It is not possible to quantitatively measure the amount of ^{240}Pu present in samples where the ^{238}Pu enrichment is 20 percent or greater. ^{240}Pu is not very abundant (about 2% of a sample enriched to 83% in ^{238}Pu), and it emits only a few gamma rays. We have developed an algorithm that predicts with sufficient accuracy the amount of ^{240}Pu present in a sample from the relative amounts of ^{239}Pu and ^{241}Pu. The user also has the option of providing the declared

^{240}Pu abundance to the data-analysis program. Also, no significant gamma rays are emitted by ^{242}Pu; hence, we cannot determine its relative abundance. However, ^{242}Pu is negligible in samples enriched to 83% in ^{238}Pu because its total abundance rarely exceeds 0.1%. The user can provide a declared ^{242}Pu abundance to the data-analysis program for completeness in the isotopic analysis.

STATUS AND RESULTS

Our first system was installed at Westinghouse Savannah River Company in August, 1990, and a second similarly designed system was delivered to the Westinghouse Hanford Company in July, 1991. The system at Hanford will measure ^{238}Pu processed into ceramic pellets approximately 2.5-cm high by 2.5 cm in diameter with an iridium cladding (referred to as fueled clads) and stored in a stainless-steel container in addition to ^{238}Pu enriched materials in EP60/61 containers. Two fueled clad pellets are placed in a stainless-steel container that is 5 cm in diameter by 10-cm tall and has a wall thickness of approximately 0.32 cm. We have not yet made measurements on this type of sample.

At Savannah River plutonium-oxide materials with ^{238}Pu abundances ranging from 20% to 85% have been measured with accuracies of better than 1% in the specific power. A precision of 0.3% on the ^{238}Pu abundance and the specific power can be obtained in an one-hour measurement of 200-grams of plutonium oxide enriched to 83% ^{238}Pu. Table 2 shows the specific power results on seven samples. Gamma-ray spectra from two samples indicated the presence of fluorine; the (α,n) interaction on ^{19}F produces ^{22}Na which gives rise to a 1275-keV gamma ray that was observed. This interaction also results in a 583-keV gamma ray that interferes with the 583-keV gamma ray from ^{228}Th decay. It was necessary to modify the data-analysis program to monitor the residual of the 583-keV peak in the fit to determine the isotopic abundances, and if the residual after several iterations exceeded five standard deviations, it is removed from the analysis.

SUMMARY

Based on our experience thus far the design of the sample enclosure and detector shield does satisfy the requirements for measuring the fissile content in these types of samples. Careful consideration had to be given in the systems' design to provide sufficient gamma-ray shielding around both the detector and its window so that the only gamma rays measured from a sample were those that pass through the window. This is necessary, so that accurate attenuation corrections can be made to the gamma-ray intensities in the final analysis. Consideration also had to be given to the

Table 2. Specific power results by gamma-ray isotopic measurement.

Declared % ^{238}Pu	Sample mass (g)	Count time (mins.)	Specific power meas. (mW/g)	Specific power meas. error (%)	Specific power % diff. from dec'ld. value
84.5	227	83.3	480.1	0.32	0.0
84.2	95	50.	477.1	0.42	-0.28
83.3	66	50.	473.5	0.42	0.17
83.3	254	133.3	472.2	0.21	-0.22
83.3	143	50.	473.4	0.35	0.05
78.7	188	50.	448.6	0.43	0.17
20.7	223	50.	124.6	1.00	-0.63

exposure of the detector to neutrons and the resulting degradation in detector performance with time. To extend lifetime of the detector, we maximized the distance between the detector and a sample and added 5 cm of borated polyethylene between them. We expect that a detector should function without significant gamma-ray peak shape deterioration for approximately one year with a sample load of three 50 minute measurements per day, *and provided no samples remain in the enclosure for long periods when not being measured.* We recommend an n-type HPGe detector, which is less sensitive to neutron damage, and can be annealed to recover its performance.[3]

The specific power results indicate that our data-analysis methodology is performing very well. The methodology uses an accurate technique based on first principles to delineate the "intrinsic" gamma-ray detection efficiency curve rather than a fit using a polynomial of some type to the data. The methodology also treats the entire set of data with consistent and appropriately weighted equations that converge to yield the isotopic abundances with rigorous errors.

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REFERENCES

1. R. GUNNINK, "MGA: A Gamma-Ray Spectrum Analysis Code for Determining Plutonium Isotopic Abundances, Volume 1: Methods and Algorithms," UCRL-LR-103220, Vol. 1, Lawrence Livermore National Laboratory, Livermore, CA (1990).

2. R. GUNNINK, W. D. RUHTER, and J. B. NIDAY, "GRPANL: A Suite of Computer Programs for Analyzing Complex Ge and Alpha-Particle Detector Spectra," UCRL-53861, Vol. 1, Lawrence Livermore National Laboratory, Livermore, CA (1988).

3. Z. KOENIG, R. GUNNINK, W. D. RUHTER, and D. C. CAMP, "Deterioration of Pu Isotopic Analysis with Neutron Damage to HPGe Detectors," UCRL-JC-105888, Lawrence Livermore National Laboratory, Livermore, CA (1991).

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