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FOUR PASSIVE ASSAY TECHNIQUES
APPLIED TO MIXED-OXIDE FUEL

by

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ABSTRACT

It was demonstrated that four different passive non-destructive assay techniques could be successfully used to measure an inventory of mixed-oxide (Pu,U)O₂ fuel rods. The techniques included: calorimetry, neutron-coincidence counting, and gamma-ray counting with NaI(Tl) detection and with an automated GeLi detection system. A thorough program of destructive measurements was also conducted, and the numerical results obtained nondestructively were compared with the destructive analyses to evaluate the reliability of the former. The comparison indicated no significant difference for three of the techniques and a difference of approximately 0.5% relative, for the neutron-coincidence-counting technique.

The inventory consisted of two loadings of approximately 12,000 and 6000 rods each and contained 15 and 30 wt %, respectively, plutonium oxide with an isotopic content of about 88% ²³⁹⁺²⁴¹Pu and 11.5% ²⁴⁰Pu.

This work demonstrated some of the advantages and limitations of each technique when applied to a real fuel-inventory measurement problem and indicated the precision and reliability that can be expected. It also made possible a collection of details of the practical aspects of actually using the techniques. Much of the information collected through the experience of this demonstration should be of direct benefit to those conducting safeguards measurements.

INTRODUCTION

Subject

This report describes the use of four different nondestructive passive assay techniques to determine the amount of plutonium contained in an inventory of nuclear-reactor fuel elements. The techniques are classified as passive since only the radiation naturally emitted by the fuel elements themselves was measured. The measurements did not require an active interrogation (or bombardment) by any external sources of radiation. The measurements did not alter the elements in any manner and hence were considered nondestructive. All measurements were made concurrently, and for most of the cases identical elements were measured by each technique. The four techniques used are listed in Table I, together with the basic measurement principle upon which each depends.

TABLE I. Passive Assay Techniques Applied to ZPR Mixed-oxide Fuel

Technique	Basic Principles of Measurement
(1) γ ray count with GeLi	Measure intensity of γ rays characteristic of isotopes of interest.
(2) γ ray count with NaI(Tl)	Measure intensity of γ rays in three spectral regions each containing energies characteristic of three isotopes of interest.
(3) Neutron-coincidence count	Measure intensity of spontaneous-fission neutrons emitted from ^{240}Pu , ^{238}Pu , and ^{242}Pu (main contribution from ^{240}Pu).
(4) Calorimetry	Measure quantity of heat generated by radioactive decay (main contribution from alpha particle emission).

The fuel consisted of pellets of the plutonium-uranium mixed-oxide variety, stacked in tubing to make the conventional fuel-rod configuration. The rods are to be used in zero-power-reactor (ZPR) type experiments. Some of the basic characteristics of this fuel are presented in Table II, and the physical appearance of the rod-type elements is illustrated by the photograph of four rods in Fig. 1.

TABLE II. Characteristics of Mixed-oxide ZPR Fuel Rods (Pellet-loaded)

Nominal dimensions:	3/8- x 6-in. SS-clad rods
Nominal weight:	89.3 g
Nominal plutonium isotopic:	^{238}Pu 0.05 wt %, ^{241}Pu 1.7 wt %, ^{239}Pu 86.55 wt %, ^{242}Pu 0.2 wt %, ^{240}Pu 11.5 wt %
Composition:	F-type rods: $(\text{U}_{0.85}\text{Pu}_{0.15})\text{O}_2$, total plutonium 13.2 wt % G-type rods: $(\text{U}_{0.70}\text{Pu}_{0.30})\text{O}_2$, total plutonium 26.2 wt %

Fig. 1. Photograph of Four Rod-type Fuel Elements. These fuel elements were nondestructively analyzed for plutonium content by the passive nondestructive technique, ANL Neg. No. 150-511.

Scope

Presented as a practical experimental approach, this report contains a limited amount of discussion of theoretical aspects. Except for the calorimetric technique, details of the apparatus design and theory of operation of the instrumentation have been covered in the published literature.¹⁻³ Specific steps followed in making the measurements are usually omitted, except where it seems appropriate to present them for purposes of clarification. The design and theory of operation of the calorimeter are described briefly since a description of the specific instrument has not yet appeared in the literature. A separate report on the design and use of the calorimeter is being prepared. Until this other report is available, readers interested in details of that instrument should contact the authors personally.

Numerical results of the measurements made with each nondestructive technique are presented together with an estimate of their precision. Results of destructive analyses, obtained by chemical and mass-spectrometric techniques, are also presented and are compared with nondestructive values in an effort to arrive at an indication of the accuracy of the latter. In addition, other factors inherent in the nature of the techniques are presented and discussed in an effort to arrive at some insight into the relative merits

of the nondestructive methods. However, it is considered beyond the scope of this report to make a final judgment upon the value of each technique.

Purpose

The purpose of this report is to describe how the four nondestructive techniques were simultaneously applied to a plutonium-fuel-inventory measurement problem and to present the information gained through this experience. Information on the following areas of interest was collected and is discussed:

1. It is desirable that the precision and accuracy attainable with these techniques, when applied to the quantitative measurement of a plutonium fuel inventory, be demonstrated.
2. Information is needed on the practical problems associated with the application of these techniques to a real fuel-inventory measurement problem.
3. The relative value of these different techniques as concerns their usefulness and reliability for various safeguards applications should be assessed.

GENERAL FEATURES OF THE MEASUREMENT

The importance of the role that each technique assumed as part of the total measurement system depended upon several things. These included: the number of measurements that could be made (i.e., the fraction of the total inventory measured), the reliability of these measurements as concerns precision and accuracy, the type of isotopes that could be detected, and the nature of the apparatus. With these criteria, gamma-ray counting with GeLi detection was assigned a major role, since the apparatus was automated, since it was sensitive to fissile isotopes, and since previous applications^{1,4,5} had demonstrated its reliability. The other three nondestructive techniques (see Table I) all assumed a secondary role of about equal importance. However, the fact that calibration of the calorimeter was not dependent upon destructive chemical analyses, as were the other three methods, added considerably to the importance of the calorimetric measurements. Concurrent with the nondestructive measurements, destructive analyses were conducted by chemical and mass-spectrometric methods and were available for comparison with the numerical results of the nondestructive measurements. Table III lists the number of measurements made by chemical and nondestructive techniques.

All rods measured were randomly selected from each production lot. The F-type rods were fabricated in nine lots and the G in four. The method

used to select rods to be assayed can be illustrated by describing the steps followed in making the selection from the group of G-type rods. The first step in selecting G rods for measurement was to randomly withdraw (using a random-number table) 20% from each of the four lots. All four of these groups were measured by the nondestructive method of gamma-ray counting with GeLi detection. In a similar manner, about 10% were then withdrawn from these four groups, and these smaller groups (i.e., about 2% of each lot) were measured in turn by the other three nondestructive assay techniques. In other words, an approximate 2% random selection was measured four times, once by each of the four different techniques, and an additional 18% measured with the GeLi system. As can be noted from Table III, 30 different samples (pellets) were chemically analyzed. These samples were also selected randomly from each production lot, and in some cases were obtained by dejacketing finished rods. Twelve of the G rods that were opened and chemically analyzed (i.e., opened and one of the 10 pellets analyzed) were available for nondestructive assay before dejacketing, and results of the measurements are included in the totals shown in Table III. The F-type rods were selected in the same manner as described above for the G type, except that the percentages were about 17 and 0.8. For the F case, it was possible to nondestructively assay 19 rods before they were dejacketed for chemical analysis. These 19 rods were evenly distributed between the last four F lots to be fabricated.

TABLE III. Number of Measurements Made by Each Technique

Fuel-rod Type:		F Rods (U _{0.85} Pu _{0.15})O ₂	G Rods (U _{0.70} Pu _{0.30})O ₂
Total Number of Rods in a Group:		12,992	6,073
Method			
γ ray count with GeLi	Number of rods measured	2,174	1,217
	% total	17	20
γ ray count with NaI(Tl)	Number of rods measured	85	133
	% total	0.7	2.2
Neutron-coincidence count	Number of rods measured	105	132
	% total	0.8	2.2
Calorimetry	Number of rods measured	107	109
	% total	0.8	1.8
Chemical	Number of samples	90	30
	% total	0.7	0.5

Two important parameters of the techniques are the measurement times and the type of isotopes measured by each. This information is listed in Table IV. The measurement times listed in Table IV are the actual number of minutes needed to measure a single fuel rod with the detection system of each technique. The length of time needed for these measurements was primarily dependent upon the sensitivity of the apparatus, the level of precision desired, and the total time available for completing the inventory verification. For these measurement times, the estimated coefficients of variation for a single rod were from 1.0% for the calorimetry to 1.4% for the neutron-coincidence count. Each technique required further processing of the data collected by this measurement in order to obtain more relevant information such as grams of ^{239}Pu and weight percent of plutonium. For the gamma-ray-counting GeLi detection method, the processing was carried out immediately following data collection, since the system had on-line computer control. Details concerning the calculations and calibrations associated with each technique are presented in the next section where a separate discussion is presented on each.

TABLE IV. Isotopes Measured by Each Nondestructive Technique and Measurement Time for a Single Rod

Passive Nondestructive Technique	Measurement Time for One Rod, min	Isotopes Measured
γ ray count with GeLi	4.5	^{239}Pu ^{241}Pu
	22.5	^{241}Am ^{238}U
γ ray count with NaI(Tl)	3.3	^{239}Pu ^{241}Pu ^{241}Am
Neutron-coincidence count	60	$\Sigma(^{240}\text{Pu}^a + ^{242}\text{Pu} +$ $^{238}\text{Pu})$
Calorimetry	80	$\Sigma(^{238}\text{Pu}^a + ^{239}\text{Pu}^a +$ $^{240}\text{Pu} + ^{242}\text{Pu} +$ $^{241}\text{Am})$

^aMain components of the sum.

Table IV indicates that a 4.5-min counting time was needed to measure the ^{239}Pu and ^{241}Pu isotopes contained in a rod. This means the system was capable of simultaneously measuring the intensity of two different gamma rays, each of which was characteristic of one of the isotopes. In a similar manner, ^{241}Am and ^{238}U were measured with a 22.5-min counting

time. A static count of 3.3 min was required for measuring the ^{239}Pu , ^{241}Pu , and ^{241}Am isotopes by gamma-ray counting with sodium iodide detection.

The isotopes measured by calorimetry or neutron-coincidence counting are listed as summations or groups of isotopes. This is meant to indicate that these detection systems responded simultaneously to radiation from a group of isotopes and could not measure radiation unique to a single isotope. However, in the neutron-coincidence-counting technique only about 3% of the total count came from the ^{242}Pu and ^{238}Pu isotopes, and hence the technique was primarily a measure of ^{240}Pu . The calorimeter was capable of measuring a quantity proportional to the rate at which heat was generated and responded to the sum of all the heat emitters, as indicated in Table IV. Since the ^{241}Am caused some heating, a correction had to be made for its contribution in order to obtain a value proportional to the total plutonium. Since the contribution from the uranium was insignificantly small, it was not included in the summation listed in Table IV. The amount of heating produced by a particular isotope contained in a fuel rod depended upon its rate and mode of decay and upon the total amount of an isotope present. The calorimetric measurements were most sensitive to those causing the most heating, and these are marked in the table as the main contributors.

Using mass-spectrometric isotope ratio data and total weights of oxide in each rod, the measurement results (i.e., quantity of a certain isotope or group of isotopes) obtained by the four techniques were converted to weight percent total plutonium (wt % Pu_T). This conversion made it possible to compare the results obtained by the different techniques with the chemical analyses, which were in terms of wt % Pu_T .

FEATURES OF THE SPECIFIC MEASUREMENT SYSTEMS

Gamma-ray Counting with GeLi Detection

As pointed out in Table I, this method is based upon the measurement of the intensity of the gamma-ray flux emitted at a particular energy which is characteristic only of a certain isotope. This is possible because of the high-resolution characteristics of the germanium-lithium drifted detector used for this measurement technique. The measurement system used for this work operates with on-line computer control and has been described several times in the literature.^{1,4,5} However, some of its main features are summarized here for completeness. The system collects and analyzes the characteristic gamma-ray intensities, accomplishes a periodic calibration, processes the data and computes isotopic content, calculates confidence bands, compares our data to data reported by the vendor, and maintains a reasonably high throughput because of the automation. Some

of the recent improvements in the method include more precise dead time and self-absorption correction techniques, which are made automatically under computer control.

As stated previously, this technique was selected as the primary nondestructive method to be used for fuel-inventory verification and was used to measure the largest number of fuel rods. Table III indicates that 2174 F-type rods and 1217 G rods were analyzed with this technique. The amount of ^{239}Pu and ^{241}Pu contained in these 3391 rods was quantitatively determined by measuring the intensity of the gamma rays having energies of 414 and 267 keV. The 414-keV energy is characteristic of the ^{239}Pu and the 267-keV energy of ^{241}Pu . This analysis was accomplished by one scan (4.5 min/rod, see Table IV) of each rod since the computer system was capable of simultaneously measuring two components.

This method was also applied to the analysis of the quantity of ^{241}Am and ^{238}U contained in this fuel. Sixty-five fuel rods (a random selection of five rods from each of the 13 different F-type production lots) were analyzed for these two isotopes. This was accomplished by measuring the gamma rays of 662-keV energy emitted by ^{241}Am and the 1.0-MeV gamma rays emitted by $^{234\text{m}}\text{Pa}$, a second daughter of ^{238}U . These two measurements were also made simultaneously with the computer-controlled system by scanning each rod five times ($5 \times 4.5 = 22.5$ min/rod). This longer counting time was necessary in order to collect a significant number of counts from these two peaks, which are of relatively low intensity. More intense gamma rays of lower energy are emitted by ^{241}Am , but the 662-keV energy was selected because it was close enough to the 1.0-MeV peak in the energy spectrum to allow a simultaneous analysis with this system. A typical gamma-ray energy spectrum obtained from this fuel using GeLi detection is shown in the upper portion of Fig. 2. The 414-keV, 662-keV, and 1.0-MeV peaks are specifically identified. The lower spectrum shown in Fig. 2 was obtained by the NaI(Tl) technique and is discussed below.

Since the apparatus used for these measurements has been previously described in the literature,^{1,4} only a few details of its construction and operation are presented at this time. The small on-line computer (PDP-8) forms the heart of the system and controls the basic components of the apparatus, such as the fuel-rod scanner-conveyor, the detector, and the analog-to-digital converters, and processes the data for immediate printout of results. Figure 3 is a block diagram of some of the main components. Figure 4 is a photograph of the scanner-conveyor table, which is loaded with fuel rods and is conveying a rod in the lower right-hand corner past the GeLi detector. The detector is shielded and collimated by the heavy cylindrically shaped lead shield which is visible in the picture.

A 45-cc GeLi detector was used, and the distance from the detector to a fuel rod was approximately 2 in. Collimation was such that the detector

Fig. 2. GeLi and NaI(Tl) γ Spectra of ZPR Mixed-oxide Fuel Rod. Energy regions and specific energy peaks useful for passive nondestructive assay are marked. ANL Neg. No. 150-524 Rev. 1.

Fig. 3

Block Diagram of Main Components of Automated Non-destructive Assay System. ANL Neg. No. 150-450.

Fig. 4. Photograph of Fuel-rod Conveyor Table and GeLi Detector. The Automatic conveyor table is covered with fuel rods which are passing under the GeLi detector in the lower right-hand corner. ANL Neg. No. 501-935.

"viewed" a rod through a $1/2$ by $1\frac{1}{2}$ -in. slot. The $1\frac{1}{2}$ -in. dimension was oriented along the length of the rod, and the $1/2$ -in. dimension viewed the full width or diameter of a rod. The lead shielding was approximately 3 in. thick in a radial direction. Figure 5 shows the small PDP-8 computer and the associated peripheral components such as teletype, tape reader, and card reader.

Fig. 5. Photograph of Small Computer and Peripherals for Automated Assay System. ANL Neg. No. 150-454.

Gamma-ray Counting with NaI(Tl) Detection

The basic principle of this technique was summarized in Table I. The technique is similar in many ways to the method reported by Bishop.^{3,5} Three regions of the gamma-ray energy spectrum, each of which contains energies characteristic of the isotopes of interest but is not resolved, are measured with a NaI(Tl) detector system. The lower portion of Fig. 2 shows a gamma-ray energy spectrum obtained from a mixed-oxide fuel rod with a NaI(Tl) detector and the three broad energy regions that were measured. The assay of plutonium by the measurement of single-energy gamma rays, as described in the previous section, is usually very difficult with NaI(Tl) because of its inferior energy resolution. This is clearly apparent when the two spectra shown in Fig. 2 are compared. However, it is possible to use intensity measurements of three relatively broad energy regions to compute the amounts of the ^{239}Pu , ^{241}Pu , and ^{241}Am isotopes contained in fuel. This is because each region contains energies characteristic of the three isotopes. The technique involves the solution of three simultaneous equations.

Contributions to the intensity of the gamma-ray flux in the three regions originate primarily from four sources: ^{239}Pu , ^{238}U , and two daughters of ^{241}Pu (^{237}U and ^{241}Am). The ^{237}U comes to equilibrium with the

^{241}Pu in a short time and can be used as a measure of the ^{241}Pu present, but the ^{241}Am is not in equilibrium and is constantly increasing. To accomplish the measurement, three regions of the NaI(Tl) gamma-ray energy spectrum were selected in the following way:

Region Designation, keV	Energy Range, keV	Main Contributor	Other Contributors
210	170-240	^{237}U	^{239}Pu , ^{241}Am , $^{238}\text{U}^*$
400	300-460	^{239}Pu	^{237}U , ^{241}Am , $^{238}\text{U}^*$
660	570-770	^{241}Am	^{239}Pu , ^{237}U , $^{238}\text{U}^*$

An equation can be written for each of the three regions by assuming that the total gamma flux contained in a region is the sum of four contributors. The equations for the regions can be written as

$$210\text{-keV region: } R = A_1X + A_2Y + A_3Z + R_u,$$

$$400\text{-keV region: } S = B_1X + B_2Y + B_3Z + S_u,$$

and

$$660\text{-keV region: } T = C_1X + C_2Y + C_3Z + T_u,$$

where

$$X = ^{239}\text{Pu content,}$$

$$Y = ^{241}\text{Pu content,}$$

$$Z = ^{241}\text{Am content,}$$

$$R = \text{count from 210-keV region,}$$

$$S = \text{count from 400-keV region,}$$

$$T = \text{count from 660-keV region,}$$

A_i , B_i , and C_i are constants determined from standard data,

and

R_u , S_u , and T_u = ^{238}U contribution to the background for each region as determined from measurement of a rod of depleted UO_2 .

*The contribution of ^{238}U is largely due to Compton scattering.

As indicated above, the constants R_u , S_u , and T_u are determined by measuring a rod of pure ^{238}U (depleted UO_2). A fraction of these counts equal to the fraction of depleted uranium contained in each rod (available from production data, destructive chemical analyses, or from nondestructive measurement with GeLi detection technique) is subtracted from the values of R , S , and T obtained from counting each rod. For example,

$$R = A_1X + A_2Y + A_3Z + R_u$$

becomes

$$R' = A_1X + A_2Y + A_3Z,$$

where

$$R' = R - R_u.$$

To obtain values for the constants A_i , B_i , and C_i , three standard rods having different isotopic ratios are measured for each energy region. Three simultaneous equations for each region can then be derived and the nine constants obtained from their solutions. For example, the three equations for the 210-keV region would be

$$R'_1 = A_1X_1 + A_2Y_1 + A_3Z_1,$$

$$R'_2 = A_1X_2 + A_2Y_2 + A_3Z_2,$$

and

$$R'_3 = A_1X_3 + A_2Y_3 + A_3Z_3.$$

Therefore, obtaining numerical values for A_i , B_i , and C_i by this calibration and measuring R' , S' , and T' for each rod, we can determine X , Y , and Z , which are, respectively, the quantities of ^{239}Pu , ^{241}Pu , and ^{241}Am contained in a fuel rod. A small computer program was prepared for the PDP-8 computer and used to make these computations. This was accomplished during periods when the computer was not being used to control the GeLi assay system.

The apparatus used for this measurement was manually operated, and a static count was made using a collimated NaI(Tl) detector which "viewed" a portion of a fuel rod. Because of the difficulty of determining a precise weight for the volume of fuel actually measured or "viewed" by the detector, calibration was done in terms of weight percent and results obtained in this form. If desired, these values could be used to compute the weight in grams of a particular isotope in a rod by using the total oxide weight as reported by the manufacturer, or reweighing and correcting for

the weight of the stainless steel tubing. Often an entire fuel element can be "viewed" (as for the other three techniques), and this can be done either by a static measurement or by scanning.

A total of 228 rods were assayed by this technique, and the results were converted to wt % Pu_T . As stated previously, the counting time used to measure a single rod was approximately 3.3 min.

The apparatus used for this work consisted of a 2 x 2-in. NaI(Tl) detector with preamplifier and shielding, a high-voltage power supply,

Fig. 6. Block Diagram of NaI(Tl) Counting System. The system was used for the nondestructive assay of mixed-oxide fuel by measuring gamma-ray intensities of relatively broad energy regions with a NaI(Tl) detector. ANL Neg. No. 150-522.

three single-channel analyzers, three scalars, a timer, and an amplifier. Figure 6 is a block diagram of this equipment arrangement. To reduce the amount of equipment, the two single-channel analyzers and two scalars could be eliminated from the system. With this change, it would be necessary to make separate measurements of each energy region instead of obtaining the three measurements simultaneously. However, the decrease in equipment cost and increase in equipment portability might compensate for this loss in efficiency. All the components needed for such a simplified system are available in single-width modules. These could be fitted into a standard six-width NIM (nuclear-instrument module) bin having dimensions of approximately $11\frac{1}{2} \times 11\frac{1}{2} \times 14$ in. and would be reasonably portable.

Before leaving the discussion of this technique, we should mention two important points concerning the nature of required calibration standards. One is that the standards should be similar to the material being analyzed, both in physical dimension and composition. If the standards differ from the unknowns, variations in self-absorption of radiation in the fuel matrix could bias the results. The second point is that the standards must have different isotopic ratios in order to obtain independent equations for determining the constants.

Neutron-coincidence Counting

It was indicated in Table I that the neutron-coincidence counting technique depends upon the measurement of spontaneous-fission neutrons. A total of 237 fuel rods, which were randomly selected by the process described earlier in this report, were measured by this technique. From the mass-spectrometric measurements of the fuel and the calibration rods, it was possible to express the measurement results in terms of wt % Pu_T .

The spontaneous-fission process, which gives rise to the simultaneous emission of two or more fast neutrons, produces neutrons at a high enough rate in the even A isotopes to be useful in the assay of ^{240}Pu . If the ratio of ^{240}Pu to the other isotopes is reliably known from other measurements, the technique can be used to determine the total plutonium. The method involves the detection of the fission neutrons by using a coincidence method² to distinguish them from the neutrons produced by (γ, n) reactions. Strain and Omohundro⁶ have reported in detail upon the type of equipment (Brookhaven version) used for these measurements. It included improvements in the system (i.e., the BNL logic) as developed by electronic scientists at Brookhaven National Laboratory as described by Sher.⁷

For specific design and operating characteristics of this equipment, the interested reader is referred to the above reports. However, for completeness, a brief description is presented here. A block diagram of the apparatus is included as Fig. 7 and may be referred to during the following discussion.

Fig. 7

Block Diagram of Neutron-coincidence Counting System. The system was used to measure the spontaneous-fission neutrons from ^{240}Pu . The apparatus included the "BNL logic." ANL Neg. No. 150-521.

fore, the quantity $R - A$ is a measure of the coincident neutrons emitted and hence the fissions occurring, which are primarily a function of the amount of ^{240}Pu present. The fast neutrons emitted by spontaneous fission must be moderated to be detectable, and a polyethylene moderator was used for this purpose, as shown in Fig. 8. Six BF_3 tubes were used as detectors and were symmetrically clustered around a central hole in the moderator where a fuel rod was inserted for counting. The polyethylene was approximately $7\frac{1}{2}$ in. in diameter and 10 in. long. The BF_3 tubes were $1\frac{1}{2}$ in. in diameter by an active length of 8 in. (1 in. beyond each end of the 6-in. fuel rods) and contained gas at a pressure of one atmosphere. As already indicated in Table II, the measurement time was 60 min. This relatively long counting time was needed in order to accumulate a sufficient number of counts (i.e., $R - A$) to

completeness, a brief description is presented here. A block diagram of the apparatus is included as Fig. 7 and may be referred to during the following discussion. After detection by BF_3 tubes, the circuitry provides output data designated as gross neutron count, real plus accidental counts (R), and accidentals (A). All pulses are recorded in the gross scaler. The count related to the number of multiple events is recorded on the real scaler, which collects pulses after a coincidence gate is activated by the first pulse of the group. The gate remains open for an appropriate time and collects real plus accidental events (R). After a delay interval long enough with respect to the neutron die-away time, a second coincidence gate opens for the same length of time, and all pulses transiting this gate are recorded on the accidental scaler as accidentals (A). Therefore,

be significant. With this counting time, the coefficient of variation for the analysis of a single rod was approximately 1.4%.

Fig. 8. Photograph of Neutron Detector-Moderator Assembly. BF_3 tubes in a polyethylene moderator surround a central hole for holding a fuel rod during measurement.

This neutron-coincidence counting technique was used for the measurement of these plutonium fuel elements for a number of reasons:

1. The technique is unique in that it is sensitive to ^{240}Pu . This allows a direct verification of the ^{240}Pu content, which is useful to reactor scientists. It is also useful to the nondestructive assayer in verifying isotopic ratios, which are needed in order to predict total plutonium content from measurements made by gamma-ray counting or calorimetry.
2. Self-absorption of the radiation is not a serious problem as with gamma-ray counting. Therefore, the technique supplies information on the plutonium content of the fuel which is independent of special corrections made for self-absorption. Sometimes this can be useful to the nondestructive assayer in evaluating the self-absorption corrections applied to the gamma-ray count data.
3. The measurement is not influenced by the presence of ^{241}Am , as are both gamma-ray counting and calorimetric techniques. Because of

this, the measurements may be helpful in determining whether there was inadequate discrimination against, or correction for, radiation emitted by ^{241}Am for the other techniques.

4. The method was useful in supporting the calorimetric measurements, since a relatively large proportion of the heat generated by this fuel came from the ^{240}Pu it contained.

5. Finally, the method was used to determine some of the practical aspects connected with actually applying the technique. These included such things as measurement times needed for this type of fuel, ease of operating the equipment, and dependability of the equipment and detector systems.

Calorimetry

Preliminary investigation indicated that measurement of the heat generated by a rod containing this type of fuel was feasible. Therefore a dry calorimeter was designed and built to accommodate fuel rods of this size and composition. Measurements were made upon 216 fuel rods, which were randomly selected from the different production lots. A single rod could be measured in approximately 90 min with a 1.0% coefficient of variation.

Basically, the calorimeter used for this work was designed to measure the wattage necessary to maintain a fuel rod at a specific temperature. This temperature was preset to a level higher than could be maintained by the heat produced by the fuel in a rod. The heat produced by a rod placed in the calorimeter sample chamber was supplemented by the heating coils surrounding the chamber until the system stabilized at the preset temperature. The total power output of a fuel rod was determined by measuring the wattage supplied by the coils to maintain a rod containing no fuel at the preset temperature and subtracting from it the wattage supplied by the coils for a rod that contained fuel. This can be restated as follows:

Measure: Y = watts supplied by the instrument to maintain a fuel rod at a temperature greater than it could maintain by its fuel alone.

Measure: W = watts to maintain a rod containing no fuel at the same temperature.

Let X = watts generated by the fuel in a rod. Then, since W is numerically equal to the $Y + X$, it follows that $X = W - Y$.

Figure 9 is a diagrammatic representation of the calorimeter sample chamber and the two concentric heat shields surrounding it. The cylindrical sample chamber and the two shields are wound with heater coils. The outer

cylinder (designated as the outer shell on the diagram) was regulated to a temperature a few degrees above ambient room temperature, the center shield was regulated to a few degrees above the outer shell, and the sample chamber regulated to a few degrees above the center shield. About 90 min after a fuel rod was inserted in the sample

A circuit diagram of the calorimeter control system is included as Fig. 10. The details of the control system and circuit design will not be discussed further at this time since they will be included in a separate report, which is currently being prepared and which is devoted to the design and application of this instrument. Since completing the measurements

Fig. 10. Circuit Diagram of Calorimeter Control System.

described in this report, we have built a second calorimeter, which reaches equilibrium in about 20 min. This new instrument, which should also be

capable of better measurement precision, will also be described in the special report referred to above.

Fig. 11. Photograph of Calorimeter Apparatus

Figure 11 is a picture of the calorimeter apparatus. The lower portion of the picture contains the control-system electronics; the rectangular box above it contains the cylinders and sample chamber. A fuel rod is inserted into the sample chamber by pushing it into the hole marked IN in the picture. It is then centered in the chamber by two plastic rods, which act as spacers and heat insulators at the ends of a rod. Also shown in the picture is a recorder for graphically displaying the output. The recording displayed in the picture indicates that the system has nearly reached equilibrium (i.e., nearly a steady output). The output can also be measured with the meter shown at the top of the picture. The calorimetric measurement of the wattage produced by the fuel was used to compute the weight percent of total plutonium (wt % Pu_T). This computation required knowledge of the

plutonium isotopic composition of the fuel (usually determined by mass-spectrometry measurements), the wt % ²⁴¹Am in the rod, the total weight of the fuel, and the power factors (i.e., watts/gram) of each isotope. The latter values can be calculated from the energies of radioactive disintegrations and the atomic mass of an isotope, or determined by calorimetric measurements of pure samples. The power factors used for computing the wt % Pu_T from the wattage measurements of this fuel were those reported by Nutter, O'Hara, and Rodenburg.⁸ The method of calculation and its derivation was as follows:

$$\text{Known: } \left\{ \begin{array}{l} \text{Total heat pro-} \\ \text{duced by a rod} \end{array} \right\} = \left\{ \begin{array}{l} \text{Sum of the quantities of heat produced} \\ \text{by each isotope contained in a rod} \end{array} \right\},$$

or

$$\left\{ \begin{array}{l} \text{Watts produced by} \\ \text{the rod as measured} \\ \text{calorimetrically} \end{array} \right\} = \left\{ \begin{array}{l} \sum [(\text{Watts from } ^{238}\text{Pu}) + (\text{Watts from } ^{239}\text{Pu}) \\ + (\text{Watts from } ^{240}\text{Pu}) + (\text{Watts from } ^{241}\text{Pu}) \\ + (\text{Watts from } ^{242}\text{Pu}) + (\text{Watts from } ^{241}\text{Am})] \end{array} \right\}^*.$$

Also known:

W = weight of the fuel (i.e., core weight or mixed oxide in this case) in grams,

I_i = weight percent of the plutonium for each plutonium isotope (determined mass spectrometrically and commonly called the "isotopic"),

J = weight percent of the core that is ^{241}Am (usually determined by chemical separation and gamma pulse analysis),

and

K_i = power factor for each isotope (watts/gram) (a constant for each isotope, which may be calculated from the energies of the disintegrations and/or by separate calorimetry measurements).

Let

C = weight percent of the core that is plutonium. (This is the measurement usually made by chemical analysis and expressed as wt % Pu_T .)

Using C, W, I_i , K_i , and J, the following relationships are derivable:

$$\text{Total grams plutonium} = W \times C;$$

$$\text{Grams of particular plutonium isotope} = I_i(W \times C);$$

$$\begin{array}{l} \text{Watts produced by the grams of a} \\ \text{particular plutonium isotope in a rod} \end{array} = K_i I_i (W \times C);$$

$$\text{Grams of } ^{241}\text{Am in a rod} = J \times W;$$

$$\begin{array}{l} \text{Watts produced by the grams of} \\ ^{241}\text{Am isotope contained in a rod} \end{array} = K_i (J \times W).$$

Therefore the original relation becomes

$$\begin{aligned} \text{Total watts measured/rod} = & \sum [K_{^{238}\text{Pu}} I_{^{238}\text{Pu}} (W \times C) + K_{^{239}\text{Pu}} I_{^{239}\text{Pu}} (W \times C) \\ & + \dots + K_{^{241}\text{Am}} (J \times W)]. \end{aligned}$$

Since all factors except the wt % Pu_T (i.e., C) are known, the equation can be solved for C.

*Watts from uranium considered too small to be significant in this measurement calculation.

With the above relation, the weight percent of total plutonium (i.e., C) was calculated for each rod that was calorimetrically measured. A short computer program was prepared for making this calculation with the small PDP-8 computer. The computer program included steps whereby corrections could be made for the decay of ^{241}Pu and growth of ^{241}Am that had occurred since the measurements of $I_{241}\text{Pu}$ and $I_{241}\text{Am}$ were made.

The usefulness of the calorimetric method of determining wt % Pu_T is limited by the fact that it requires reliable measurements of the isotopic ratios, core weight, and power constants of the isotopes. However, it is almost totally independent of any calibration based upon a chemical analysis. This is an important and unique feature of the method, since the calibration of the other three techniques depends upon a chemical analysis of "standard calibration rods" in order to obtain an expression relating counts to quantity of plutonium. The difficulties associated with obtaining precise and accurate chemical analyses of calibration standards are avoided. It is also worth noting that the wattage measurements depend only upon an electrical calibration, which can usually be done with greater precision than most chemical analyses. Sometimes pure wattage measurements can be useful. Often the net weight (i.e., weight of mixed oxide) of fuel rods is accurately known. If this is the case, the watts/gram can be determined with high precision, since this value depends only upon the electrical calibration and the weight measurement. These precise measurements of watts/gram can sometimes be useful in detecting small relative differences in the total isotopic content of rods. Of course, relative measurements of this type tell nothing about the particular isotopes responsible for any differences detected.

MEASUREMENT RESULTS

Single Rods

Individual rods were measured when applying these techniques, and the number of grams of each isotope to which a method was sensitive was determined. Table V illustrates the numerical results obtained by

TABLE V. Typical Measurement Results for a Single Fuel Rod^a

Method	Components Measured, g					
	^{239}Pu	$^{240}\text{Pu} +$ $^{241}\text{Pu} + ^{242}\text{Pu}$	^{241}Pu	^{241}Am	^{238}U	Pu_T
γ ray count with GeLi	20.30		0.41	0.044	54.45	23.47
γ ray count with NaI(Tl)	20.34		0.40	0.040		23.52
Neutron-coincidence count		2.80				23.39
Calorimetry						23.65
Destructive (Chemical, mass spectrometric)	20.35	2.82	0.40	0.040	54.52	23.53

^aResults are listed in terms of the average grams/rod of the different components for a G-type ($\text{U}_{0.70}\text{Pu}_{0.30}$) O_2 rod from lot 12. The destructive analyses are also indicated for comparison.

the nondestructive measurement of single rods, together with the results obtained destructively from chemical and mass-spectrometric measurements for comparison.

Groups of Rods and the Total Inventory

The individual analyses of single rods were summed to obtain the total quantity of a particular isotope contained in each production lot and then converted to weight percent total plutonium. Expressing our measurement results in terms of weight percent total plutonium for each lot was the form most useful in verifying the total fuel content of the inventory. This was because the fabricator made a new oxide blend for each lot and reported his chemical analysis (i.e., wt % Pu_T) of each as representing the quantity of fuel he supplied. Our chemists also made measurements of each lot by randomly selecting rods from each lot and destructively analyzing them. Therefore, two chemical (destructive) analyses, four nondestructive measurements, and the production data were available for comparison on a lot-by-lot basis to determine the fuel contained in the total inventory. As pointed out previously, the F-type ($U_{0.85}, Pu_{0.15}$) O_2 group of rods consisted of nine lots. The measurement results for the entire group (i.e., sum of nine lots) are presented as a bar graph in Fig. 12. Figure 13 is a similar bar graph of measurements for G-type ($U_{0.70}, Pu_{0.30}$) O_2 rods, except that results for individual lots are also included with results for the entire group. The results obtained for the entire inventory are summarized in Table III. The cross-hatched areas at the top of the bars in Fig. 12 indicate an approximation of the precision of each technique.

Fig. 12. Bar Graph of Measurement Results from All Techniques for F-type ($U_{0.85}, Pu_{0.15}$) O_2 Rods. ANL Neg. No. 150-525.

Fig. 13. Bar Graph of Measurement Results from All Techniques for G-type ($U_{0.70}, Pu_{0.30}$) O_2 rods. ANL Neg. No. 150-523 Rev. 1.

SUMMARY AND CONCLUSIONS

An important goal of the project was to nondestructively verify the quantity of fuel contained in this inventory. We were particularly interested in obtaining nondestructive measurements of this fuel, since a thorough program of destructive analyses was carried out concurrently and would allow an evaluation through the quantitative comparison of numerical results. The quantity of fuel determined by the two gamma-ray techniques and by calorimetry agree with the chemical measurements, since differences between the numerical results (see Table VI) are small and lose significance when the estimates of precision are considered. Only for neutron-coincidence counting is it possible that a real difference may exist, and this is on the order of only 0.5% relative.

TABLE VI. Summary of Results of Nondestructive and Destructive Assays of Mixed-oxide Fuel Inventory

Method	Type of Fuel: F Rods ($U_{0.85}Pu_{0.15}$)O ₂			G Rods ($U_{0.70}Pu_{0.30}$)O ₂		
	Total Inventory Size: 12,992 Rods			6073 Rods		
	Sample Size		Results, wt % Pu _T	Sample Size		Results, wt % Pu _T
	No. of Rods	% Total		No. of Rods	% Total	
γ ray count with GeLi	2174	17	13.28 ± 0.03	1217	20	26.42 ± 0.04
γ ray count with NaI(Tl)	85	0.7	13.22 ± 0.02	133	2.2	26.62 ± 0.02
Neutron-coincidence count	105	0.8	13.35 ± 0.06	132	2.2	26.26 ± 0.07
Calorimetry	107	0.8	13.20 ± 0.03	109	1.8	26.51 ± 0.03
Chemical	90	0.7	13.19 ± 0.02	30	0.5	26.48 ± 0.04

An evaluation based entirely upon a comparison of numerical results is useful, but should not be the only criterion used in selecting a method or group of methods. Experience with this study made it evident that other factors connected with the nature of the technique and the conditions under which it must be applied should be considered. Some of the advantages and limitations of using a particular technique or several simultaneously are presented below with a separate discussion of each.

This work reaffirmed that the gamma-ray counting technique using GeLi detection is a reliable method of nondestructively measuring ²³⁹Pu and ²⁴¹Pu. In addition, it also showed that the quantities of ²⁴¹Am and ²³⁸U can be reliably determined. And it was demonstrated that the method can be used for the accurate determination of the total plutonium when reliable mass-spectrometric measurements of the isotopic ratios are available. It is probably the best choice of the four if a single method must be used to measure this type of fuel inventory. The results indicated that reliable corrections for self-absorption and dead time can be made, that accurate calibration is possible, and that the counting times are reasonably short. Once the system parameters are determined and a good computer program

established, fuel can be measured rapidly enough so that a large part or even the total inventory can be assayed.

When the work can be done at a permanent laboratory installation, as in this case, the equipment complexity and the lack of portability associated with using on-line computer control, a GeLi detector, and a large conveyor system pose no problem. However, although the computer and large scanner were valuable, gamma-ray counting with GeLi detection can be and in many cases is usefully applied without such equipment. Many times, particularly for small inventories, the use of an on-line computer for data processing and control of a large scanning table is unnecessary.

One of the most important characteristics of the gamma-ray counting technique with GeLi detection is that it is the only one of the four methods that directly measures the quantities of fissile components of the fuel. This is possible because gamma rays that are characteristic only of a particular component of interest can be measured with the GeLi detection system. The other techniques are based upon indirect measurements since their detection systems respond simultaneously to a group of components or only to non-fissile components as in the case of the neutron-coincidence counting. When the response is indirect, other measurements, which were obtained destructively or by another nondestructive test, must be used to convert the original data to a more useful form. This conversion introduces sources of error and usually results in greater uncertainty.

The gamma-ray counting technique using NaI(Tl) detection might be the preferred method if short measurement time and relatively simple, inexpensive, and portable equipment are important factors. This could be the case for a visiting safeguards inspector. However, the data reduction required for this method (e.g., simultaneous equations and solution) could present problems or delays when computing equipment is not readily accessible. Also, as mentioned during the discussion of this method, the selection of adequate calibration standards appears to be a critical problem and is probably not completely understood. Further study of this problem may clarify the situation.

The neutron-coincidence counting method appears to be a useful technique for measuring ^{240}Pu . If reliable mass-spectrometric measurements of isotopic ratios are available, as was the case for this work, this method can also be used to determine the amount of fissile components or the total plutonium. The counting times required for measuring a single rod of this type of fuel were relatively long (e.g., 1 hr or more). However, it was discovered that a simple alteration of the detector-moderator assembly could be made which would allow the simultaneous measurement of seven or eight rods. This would substantially reduce the total measurement time required. Direct supportive data are not available at this time, but it appears that the measurement precision of a total inventory would not be

seriously affected if rods are measured in groups of seven or eight. An advantage of the technique is its insensitivity to radiation self-absorption. Special corrections for self-absorption, which are usually required for gamma counting, are not as critical and are often unnecessary. For this reason, the technique is particularly valuable when the fuel matrix or geometrical configuration is variable. In such cases, precise self-absorption corrections of the type required for accurate gamma-ray counting are difficult if not impossible.

Experience with the calorimetry technique showed that it could be used to obtain precise and accurate measurements of the total plutonium content of mixed-oxide fuel rods. Of course, this was possible primarily because highly reliable mass-spectrometric analyses and weights of oxide were available. If reliable isotopic data cannot be obtained, the method is of questionable value. However, good mass-spectrometric measurements are available many times when chemical analyses are missing or of uncertain reliability. The technique does not require self-absorption corrections and the equipment is relatively simple to operate. Calorimeters are often large and difficult to move, but the particular instrument used for this demonstration was reasonably portable.

A unique and valuable characteristic of this technique is that the calibration can be done electrically and is independent of chemistry. It does not depend upon standards that were chemically analyzed as with the other three techniques. This presents the opportunity to use it to verify the reliability of the chemical analyses of the rods used as calibration standards with the other methods. Usually the relatively long measurement times (90 min) required for this calorimeter approached practical limits. Experience with this instrument suggested some changes in the design which could shorten the measurement time. As a result, a new calorimeter was built which was designed to require measurement times of about 20 min. This instrument, which is also portable, is presently being evaluated.

In conclusion, probably the most important results of this project were a collection of details concerning the practical details of applying the techniques. Experience as consultants on the safeguards plant-instrumentation program indicated that potential users of nondestructive measurement techniques needed information on the practical aspects. Even though basic principles might be reasonably well understood, progress in the application was sometimes slow. It was felt that the most relevant information of this type could be obtained from the experience of demonstrating how the methods could be applied to the solution of a real fuel-inventory measurement problem. The work resulted in the collection of many details of this type, such as counting times, isotopes measurable, data reduction and calibration techniques, sample size and selection, and apparatus requirements. These are presented in the tables and in the descriptions of specific procedures in this report. Groups undertaking nondestructive fuel-measurement programs for safeguards or other purposes should find much of this information useful.

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