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JOINT PLANT INSTRUMENTATION PROGRAM AND
INTEGRATED SAFEGUARDS EXPERIMENT

FINAL REPORT

D. C. WADEKAMPER, D. M. BISHOP
AND L. T. HAGIE

U. S. ATOMIC ENERGY COMMISSION
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compiled by

D. C. Wadekamper, D. M. Bishop, and L. T. Hagie

Approved:

E. F. Kurtz, Project Manager

H. H. Klepfer, Manager
Development Engineering

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Compiled by
D. C. Wadekamper, D. M. Bishop, and L. T. Hagie

ABSTRACT

Mixed (uranium-plutonium) oxide standards were prepared and utilized to qualify available nondestructive assay equipment. Equipment was evaluated during normal operation in a plutonium facility. Measured material balances were calculated for two separate fabrication campaigns. Plutonium material unaccounted for values of less than 0.5% based on the total throughput were obtained in the final balance. A special test was undertaken to compare the plutonium content of single pellets determined by available nondestructive assay methods with controlled potential coulometric values. The implications of both programs on safeguards are summarized.

I. INTRODUCTION

The General Electric Company entered into a contract with the United States Atomic Energy Commission, Contract Number AT(30-1)-68, to support technical activities associated with the Plant Instrumentation Program (PIP) and the Integrated Safeguards Experiment (ISE). The programs were sponsored by the Office of Safeguards and Materials Management and administration was provided by the Brookhaven Office. The definition of program technical objectives as well as provisions for necessary equipment and technical direction were administered through the Brookhaven National Laboratory – Technical Support Organization.

The PIP was an experimental program directed toward the demonstration and evaluation of nondestructive techniques for the assay of nuclear material. Over-all program objectives were formulated to result ultimately in the development of a technological basis for a complex system of nondestructive instruments intended to aid in safeguarding high strategic value materials. At the same time, the equipment provided in-plant inventory verification after certain processing operations that might render past measurements doubtful or invalid.

The purpose of the ISE was to evaluate the usefulness to safeguards of material balance techniques based completely on measurements. Normal production and product certification data for nuclear material were coupled with information obtained from nondestructive assay measurements accomplished with equipment available from the General Electric Company portion of the PIP. The necessary material balance information was compiled under actual fuel fabrication conditions during two processing campaigns. Over-all program objectives were formulated to demonstrate to the nuclear industry the usefulness of material balance accounting for safeguarding plutonium.

During part of the contract period, either PIP or ISE personnel were on-site at the General Electric Company, Vallecitos Nuclear Center, Pleasanton, California. These personnel, ISE – Technical Representatives, worked jointly with General Electric personnel during planning and data evaluation of measurements and experimental tests. The ISE – Technical Representatives were specialists in the safeguards field with experience in nondestructive instrumentation and material balance calculations. Personnel from the following organizations were involved: United States Atomic Energy Commission, Brookhaven National Laboratory, National Bureau of Standards, and the International Atomic Energy Agency.

II. SUMMARY

The results of these programs indicated that material balance accounting based on measurements can be successfully utilized to detect plutonium loss or diversion during routine fuel fabrication. It was established that the desired accuracy could be met if the uncertainties in the measurements in the feed and product streams were minimized and the scrap and waste discard streams were measured with nondestructive assay (NDA) techniques.

Typical mixed (uranium-plutonium) oxide standards were fabricated and used to standardize and qualify the following nondestructive assay techniques: (1) sodium iodide and germanium (lithium) gamma spectroscopy, (2) gross and coincident neutron counting, and (3) twin isothermal calorimetry. The random measurement uncertainties ranged from a low of 0.1% for gross neutron measurement of plutonia feed to a high of 30% for coincident neutron measurement of plutonium bearing waste discards. The effects of the nondestructive assay techniques on material balance accounting were determined by measuring feed, product, scrap, and waste-discard material streams.

The two material balances prepared from historical data resulted in plutonium material unaccounted for (MUF) values of 1.54 and 0.46%. As a result of the balances, the normal operating procedures and data forms were modified to provide the necessary inputs to calculate measured material balances for the two ISE campaigns.

Three different types of material balances were calculated during ISE Campaign 1. The first type, a trial material balance, was representative of measured material balance techniques before the ISE. The material balance resulted in plutonium MUF values of 1.40 and 1.74% for enrichments 1 and 2, respectively, which were larger than the associated limits of error (LEMUF) quantities. The second balance, NDA Balance 1, was prepared similar to the trial balance with the exception of waste-discards and some scrap material streams which were defined by NDA measurements. The balance resulted in plutonium MUF values of 0.89 and 1.41% for enrichments 1 and 2, respectively, which were larger than the associated LEMUF quantities. The third balance, NDA Balance 2, utilized the best available measurements for each material stream. The plutonium MUF values of 1.14 and 1.62% for enrichments 1 and 2, respectively were smaller than the associated LEMUF quantities.

Two material balances were prepared for ISE Campaign 2. The trial material balance resulted in plutonium MUF values of 0.67 and 0.67% for enrichments 1 and 2, respectively, which were larger than the associated LEMUF quantities. The plutonium MUF values for NDA Balance 1 were 0.24 and 0.10% for enrichments 1 and 2, respectively. These MUF values were both smaller than the associated limits of error.

Comparison of the material balances for ISE Campaigns 1 and 2, normalized to ISE Campaign 2 throughput rates (~ 2500 grams plutonium), indicated that the majority of the reduction in the contributions to plutonium MUF for ISE Campaign 2 resulted from the elimination of a bias in the plutonia feed measurement. An additional reduction in the contribution to plutonium MUF resulted from measurement of heterogeneous scrap and waste-discards by non-destructive assay techniques.

A special test (ISE-2 Nested Design) was completed which compared the plutonium content of single pellets determined by sodium iodide gamma spectroscopy, gross neutron, and coincident neutron counting with data obtained from controlled potential coulometry. The random measurement uncertainties ranged from 0.3% relative for coulometry to 0.8% relative for the nondestructive assay techniques.

A comparison of completed work with the original scope of the contract is included. The over-all implications of these programs on safeguards are summarized.

III. NONDESTRUCTIVE ASSAY MEASUREMENTS

A. MEASUREMENT SUMMARY

1. General Applications

The application of nondestructive nuclear assay instrumentation to plutonium safeguards materials control has been under intensive development as a direct measurement method to provide both materials balance and diversion surveillance information. The PIP and portions of ISE represented an evaluation of promising nondestructive assay techniques by private fuel processors under in-plant conditions. As part of these programs, typical mixed (uranium-plutonium) oxide fuel rod, scrap, and waste-discard standards were fabricated; available assay equipment and techniques were qualified; and the usefulness and acceptability of selected assay techniques as a material balance tool were determined.

Nondestructive assay equipment and techniques evaluated included:

- a. Sodium iodide and germanium (lithium) gamma spectroscopy,
- b. Gross and coincident neutron counting, and
- c. Twin isothermal calorimetry.

From both an accountability and fabrication process control standpoint, feed material "inputs" and fuel rod, scrap, and waste-discard "outputs" were the initial candidates for applications of nondestructive assay techniques. Quantitative plutonium analyses were the primary goal in each case. Initial work (Standardization and Qualification) was directed toward the general application of assay techniques to a broad spectrum of fuel standards to evaluate and qualify each assay method for different measurement problems. A representative sampling of available plutonia feed material isotopics and mixed (uranium-plutonium) oxide blend enrichments were selected for fuel standards during the Standardization and Qualification phase. The standards included both 80 and 90% fissile plutonia in plutonium enrichments up to 10%. The most appropriate techniques were then selected for use during actual fuel rod fabrication projects. This simulated production application (Fabrication Applications) was accomplished on known isotopic lots of multiple enrichment fuel during the ISE campaigns. A general description of nondestructive methods utilized for both Standardization and Qualification as well as Fabrication Applications is outlined in Table 1.

TABLE 1. Nondestructive Assay Test Plan
(X Indicates Areas of Evaluation)

	GAMMA SCAN		NEUTRON		CALORIMETRY	
	<u>NaI</u>	<u>GeLi</u>	<u>Gross</u>	<u>Coincident</u>	<u>Macro</u>	<u>Micro</u>
STANDARDIZATION AND QUALIFICATION						
Rods	X	X	X	X		
Scrap	X		X	X	X	
Waste-Discards	X		X	X		
FABRICATION APPLICATIONS						
Feed Material			X	X	X	
Process Samples	X		X	X		X
Rods	X	X				
Scrap			X	X		
Waste-Discards			X	X		

It is important to note that in several instances during these programs, limited equipment availability required the use of instrumentation on applications beyond original design capabilities. For example, a 1-gallon capacity neutron coincident well count was used for single pellet analyses. In another case, a 1-watt capacity calorimeter was used to measure milliwatt heat outputs from single pellets. All such measurements contributed to the definition of minimum equipment design thresholds. However, test data resulting from such nonoptimized instrumentation should not be considered typical of ultimate measurement capabilities. Instrumentation designed for specific measurement requirements should yield superior results.

2. Measurement Basis

a. Gamma Sodium Iodide

A gamma scan method was developed using a sodium iodide detector and a multi-channel analyzer to measure characteristic gamma radiation naturally emitted from mixed oxide fuel. A data stripping technique was developed to utilize appropriate fuel standards and to compare isotopic channel contribution ratios to quantify typical Am-241, U-237, and U-238 sources of interference within the characteristic 0.3- to 0.5-MeV plutonium spectral region. Four isotopes (Pu-239, Pu-241, Am-241, and U-238) were used in the four spectral channel technique. Within the limiting assumptions of the technique, the count rate in each of the four channels with a given sample was used to define four simultaneous equations with four unknowns. The resultant equations were solved for the mass of each of the contributing isotopes.⁽¹⁾ Rod scanning was accomplished in the horizontal position by pulling the fuel rods past a shielded detector (Figure 1). An integral count was taken for each rod at a scan speed of approximately 1 ft/min. Process standardization was performed by using one rod of each fuel type and physical form as a control. All PIP standard and ISE Campaign 1 and 2 fuel rods were gamma scanned using this technique.⁽²⁾

FIGURE 1. ROD SCAN FIXTURE

The sodium iodide gamma counting of fuel scrap standards was accomplished by rotating samples in a shielded well counter.⁽³⁾ Promising passive neutron test results for fuel scrap standards resulted in the discontinuance of work in the gamma counting area.

b. Gamma Germanium (Lithium)

A method was developed for gamma scanning fuel rods using a 50-cc germanium (lithium) detector and a multi-channel analyzer (Figures 2 and 3, respectively). The technique involved integrating the 0.208-MeV peak resulting from U-237 (a daughter of Pu-241) and the 0.414-MeV complex of peaks resulting from Pu-239. All ISE Campaign 1 and 2 fuel rods were scanned with this technique.⁽⁴⁾ Gamma germanium (lithium) spectroscopy was not used for analyses where self-absorption effects posed a serious problem. Thus, scrap material analyses were performed using passive neutron counting methods. Timing limitations did not allow development of procedures to utilize Ge(Li) methods for waste-discards assay. However, the method has advantages worthy of future development for such applications.

FIGURE 2. GERMANIUM (LITHIUM) DETECTOR

c. Gross and Coincident Neutron Counting

Passive neutron counting instrumentation was supplied by the Brookhaven National Laboratory — Technical Support Organization. It consisted of the 4π geometry well counter (Figure 4) made up of a cylinder of polyethylene moderating material with a centrally located sample cavity covered with a removable plug. This cavity was surrounded by two concentric arrays (12 each) of $B^{10}F_3$ gas proportional counters embedded in the moderating material. The moderator thermalized fast neutrons to allow more efficient capture by the $B^{10}F_3$ counters. Over-all capture efficiency⁽⁵⁾ was on the order of 8%.

FIGURE 3. MULTICHANNEL ANALYZER INSTRUMENTATION

FIGURE 4. GROSS AND COINCIDENT NEUTRON ASSAY STATION

Fast neutrons may be emitted by spontaneous fission or by (γ, n) and (α, n) reactions with light elements. The principal source of spontaneous neutrons in mixed oxide fuels is Pu-240. It was necessary to independently measure the quantity of spontaneously fissioning isotopes present to predict the amount of total plutonium or thermally fissionable material in a test sample. Thus, passive counting techniques rely on the previous knowledge of sample material isotopic content. Two passive neutron counting techniques were tested: (a) Gross neutron counting where the count data are proportional to the total number of spontaneous neutrons emitted by spontaneous fission as well as neutrons emitted by (γ, n) and (α, n) reactions, and (b) Coincident neutron counting where (γ, n) and (α, n) neutron contributions are eliminated and result in count rates which are a direct function of the spontaneous fission rate of the material.^(6,7)

Standardization and qualification tests were performed on rod, scrap, and waste-discard fuel standards by using both passive assay techniques. The methods were also used for ISE feed, process sample, scrap and waste-discard analyses. The ISE rod scanning was not accomplished by neutron counting because of concurrent use of the equipment for waste-scrap analyses and technical difficulties associated with modifying the available well counter to yield satisfactory scan results. However, a similar passive neutron counting system has been successfully used for rod scanning.⁽⁸⁾

d. Calorimetry

Twin isothermal calorimetric equipment supplied by Mound Laboratory is shown in Figures 5 and 6. A 5-watt capacity macrocalorimeter was used to measure 1 kg lots of as-received plutonia powder. A second 1-watt capacity microcalorimeter was used to measure mixed oxide fuel materials in the milliwatt heat output range.⁽⁹⁾ The output data from each calorimeter were in terms of watts. This is an absolute measurement; that is, the heat from the sample is measured and compared to a known, measured amount of electrical heat. Several factors contribute to assay calculations based on these wattage data:

1. Isotopic abundance of plutonium,
2. Americium concentration,
3. Heat outputs per gram of isotope, and
4. Half-life data for the isotopes.

The last two items are known to a great degree of certainty. The first two must be determined analytically.

FIGURE 5. CALORIMETER ASSAY EQUIPMENT STATION

FIGURE 6. MACRO AND MICRO CALORIMETER

B. MEASUREMENT ERRORS

1. Precisions and Intercomparisons

A summary of Standardization and Qualification as well as Fabrication Applications precisions is presented in Table 2. Details of the experimental methods have been previously reported.⁽¹⁰⁾ Specific results for each method are compared to normal accountability data in the following sections. In evaluating these data, it is important to recognize that there are certain areas where nondestructive measurement capabilities do not compare favorably with analytical chemistry results. Typical examples are gamma sodium iodide and germanium (lithium) rod scan techniques (both $\pm 1\%$ precision) compared to a $\pm 0.2\%$ capability for analytical chemistry. However, the 100% inspection of the nondestructive techniques are especially advantageous. In many safeguards applications, this attribute overshadows the apparent precision of the traditional analytical method.

TABLE 2. Relative Nondestructive Assay Precisions at 95% Confidence

	GAMMA SCAN		NEUTRON		CALORIMETRY	
	NaI	GeLi	Gross	Coincident	Macro	Micro
STANDARDIZATION AND QUALIFICATION						
Rods	2%	2%	2%	2%	—	—
Scrap	25%	—	5%	3%	10%	—
Waste-Discards	50%	—	30%	20%	—	—
FABRICATION APPLICATIONS						
Feed Material	—	—	0.1%	0.5%	4%(a)	—
Process Samples	1 to 3%	—	5%	2%	—	5%
Rods	1%	1%	—	—	—	—
Scrap	—	—	5%	2%	—	—
Waste-Discards	—	—	20%	30%	—	—

(a) 0.03% wattage reproducibility

a. Gamma-Sodium Iodide

Sodium iodide scan results for the 48 ISE Campaign 1 and 2 fuel rods showed 6126.30 ± 67.16 grams of total plutonium at 95% confidence (1.1% relative). Analytical chemistry data for the same rods were 6103.4 ± 12.66 grams of total plutonium at 95% confidence (0.2% relative). Limits of error for rod scan test results were empirically determined and do not contain a systematic component.⁽¹¹⁾

b. Gamma-Germanium (Lithium)

Germanium (lithium) rod scan results were analyzed by ISE — Technical Representatives. Counting statistics on the order of 0.5 to 1% relative were observed. A comparison of these data with sodium iodide rod scan results has been published.⁽¹²⁾

c. Neutron Counting

Both gross and coincident neutron methods showed satisfactory results on homogeneous as-received plutonia feed material.⁽¹³⁾ Average deviations from analytical chemistry data on 11 individual 1-kg size cans of plutonia were 0.1 and 0.5%, for gross and coincident counting, respectively. With cadmium poisoning, $B^{10}F_3$ detector response was linear for samples up to 200 grams of Pu-240 equivalent. The unpoisoned system showed nonlinearity resulting from self-multiplication effects on samples containing more than approximately 50 grams of Pu-240 equivalent.

Passive neutron methods for heterogeneous scrap and waste-discard assay determinations were extremely successful. Test precision on the order of 2 to 20% were observed for scrap and waste-discard materials.⁽¹⁴⁾ The determination of systematic sources of error was not possible because available fuel standards were not verified on an absolute basis.⁽¹⁵⁾

The over-all ramification of the use of neutron counting for waste assay is best shown by its effect on typical material balance data (Table 3). Normal methods of accounting are compared to passive neutron data for typical fuel measurements. As expected, no significant differences were observed for homogeneous materials, such as clean scrap. The smaller limit of error associated with the analytical chemistry data for clean scrap makes it the preferable method for establishing material balance values. However, the use of the same procedure for heterogeneous materials (dirty scrap as an example) resulted in approximately 10% overestimates of the assay value. This lack of agreement resulted from the inability to obtain representative samples because of foreign material. Waste-discard materials were underestimated by a factor of 2 to 4 as a result of "estimating" procedures. Exhaust filters were grossly underestimated for the same reason. It is difficult to establish meaningful limits of error for "estimated" values. The important point is that the larger relative errors of the nondestructive counting techniques as compared to analytical chemical methods were less pronounced in applications where the sampling error was very large. For example, in the assay of waste material, it may be more accurate to count an entire waste container than to precisely measure several nonrepresentative samples.

TABLE 3. Typical ISE Material Stream Comparison*

	Analytical Chemistry			Neutron Assay		
	TOT Pu(g)	LE(g)	LE%	TOT Pu(g)	LE(g)	LE%
Homogeneous — Clean Scrap	214.2	0.85	0.4	213.7	5.61	2.6
Heterogeneous — Dirty Scrap	10.7	0.04	0.4	9.8	0.64	6.5
— Waste Discards	4.36**	—	—	11.4	0.04	3.5
— Exhaust Filters	0.01**	—	—	6.0	2.18	36.5

*data extracted from Table 1, GEAP-12114-3, Page 5

**estimates

d. Calorimetry

Test precisions based on wattage data for 1-kg lots of plutonia feed material were on the order of 0.03% relative at 95% confidence.⁽¹⁶⁾ Propagated errors for total plutonium contents of 4.3% relative and 7.8% relative for 90 and 80% fissile plutonium, respectively⁽¹⁷⁾ resulted primarily from analytical uncertainties for Pu-238 ($\pm 10\%$) and Am-241 ($\pm 27\%$). This highlights the need for improved analytical methods to provide full advantage of current calorimetric measurement capabilities for safeguards measurements.

Useful absolute calorimetric measurements require plutonium and americium isotopic analyses which extend beyond current industrial capabilities. The cumulative heat output of Pu-238 and Am-241 is 25 and 45% of the total wattage value for 90 and 80% fissile plutonium, respectively. However, it does appear feasible to utilize calorimetry on a routine basis for many applications, such as the evaluation of plutonia shipper-receiver differences. A simplified shipper-receiver procedure is outlined as follows:

1. The sealed container of plutonia would be calorimetered by the "shipper." The test output would be a wattage signature of the container. The increase in heat output resulting from Am-241 in-growth would be plotted versus time.
2. The sealed container of plutonia would be calorimetered again by the "receiver" with similar instrumentation. These data would also be plotted versus time using the original shipper analysis date as time zero.

The agreement between the two measurements (adjusted for decay heat) would assure the correct number of watts of material had been transferred. The result would be a quantitative basis for accepting or rejecting the shipment. Since

the container would not have been opened, resolution of shipper-receiver problems should be minimized. A simulated shipper-receiver calorimeter sequence is presented in Figure 7 for a 1-kg can of plutonia. Eight calorimetric measurements made over a 30-day period were arbitrarily divided to simulate shipper versus receiver measurements. Calorimeter test results based on wattage data were $\pm 0.03\%$ (precision). Analytical chemistry methods of evaluating shipper-receiver differences would be $\pm 0.2\%$ relative without considering sampling errors.

Test results on mixed oxide fuel scrap standards containing 10- to 20-gram plutonium in the macrocalorimeter and mixed oxide pellet standards containing 1- to 5-gram plutonium in the microcalorimeter yielded 5 to 10% accuracy, respectively.⁽¹⁸⁾ These results were not considered representative of calorimetric system capabilities because of the use of a nonoptimal calorimeter design for the wattage ranges measured. These data pointed out the need for limiting the use of calorimeters to specified heat output ranges. As an example, a milliwatt range calorimeter is capable of measuring 10 to 20 grams of plutonium scrap to within $\pm 0.5\%$.

C. FABRICATION APPLICATIONS

1. Equipment Stability and Reliability

The over-all operating dependability and stability of instrumentation tested as part of PIP/ISE was satisfactory. The calorimeter was the most stable system because of its internal method of electrical calibration and the stabilizing effect of the controlled temperature water bath. Statistically significant time dependencies were encountered with both the passive neutron and gamma counting systems. However, drift problems were satisfactorily eliminated by periodically running appropriate standards and normalizing test data within short-term periods.

The modular nature of most instrumentation greatly facilitated rapid equipment repair with minimal equipment down-time.

2. Standardization

The selection and characterization of an appropriate range of fuel rod, scrap and waste-discard standards proved vitally important during PIP/ISE. Measurement requirements must be bracketed by a range of standards typical of the physical and chemical form of the materials to be analyzed.

The lack of absolute measurements on available PIP standards precluded the establishment of systematic errors for PIP/ISE nondestructive assay measurements.⁽¹⁹⁾

3. Data Reduction and Modeling

The reduction of nondestructive assay test data was in general computerized to facilitate the handling of bulk data and lengthy limit of error calculations. For example, computerized data reduction models were developed for gamma rod scanning⁽²⁰⁾ and passive neutron counting.⁽²¹⁾ Time share, batch processing and "mini"-computers were used to reduce, evaluate, and report test data in a format suitable for subsequent use. It should be noted that no standard statistical techniques were commonly available for treating nondestructive assay test data. For example, joint ISE — Technical Representative and GE discussions resulted in as many as four independent and acceptable statistical models for the reduction of scrap and waste-discard neutron counting data (each of which produced slightly different limits of error).⁽²²⁾ This emphasizes the need for a mutually acceptable and standardized method for the statistical treatment of such data.

IV. MATERIAL BALANCE CALCULATIONS

A. HISTORICAL CAMPAIGNS

During the initial phases of the ISE, data accumulated during two previous fabrication campaigns (Campaigns A and B) were examined to determine its applicability to material balance safeguards. The ISE — Technical Representatives and General Electric personnel considered four different methods of constructing material balances from the historical data.

*All measurements ± 0.0012 watts ($\pm 0.03\%$)

FIGURE 7. CALORIMETRY OF PLUTONIA

The material balances could have been constructed in terms of (a) total oxide (including urania, plutonia, or combinations of both), (b) total metal, (c) uranium, or (d) plutonium. During evaluation of the historical data material balances were prepared by at least three different methods. The basic method was a total oxide balance which utilized both measured and estimated oxide values obtained during routine fuel fabrication. A second balance was prepared by applying theoretical metal concentration factors as well as the nominal target value for plutonium enrichment to the total oxide balance. The third balance utilized the normal process control and product certification data to obtain actual measured values of plutonium for many of the streams in the total oxide balance.

The historical material balances were prepared by applying theoretical metal concentration values as well as the nominal target value for plutonium enrichment to the total oxide balance (second balance method). This method of calculation was also used to maintain accountability control during actual processing. Material balance calculations, made by using the shipper's value for plutonium assay, resulted in a plutonium MUF value of 1.54 and 0.46%, for Campaign A and B, respectively.⁽²³⁾ A summary of the material balance results is presented in Table 4. The large MUF values were caused by inaccurate measurement of the plutonia feed because of moisture adsorption⁽²⁴⁾ and unmeasured (estimated) or difficult to measure scrap and waste streams.

Although some of the measured quantities could be associated with limits of error, this was not true for all the values. Some of the quantities in the material balance were measured, others estimated, while still other values were obtained by difference. As a result, even if meaningful limits of error were available for all other measured quantities, the material balance limit of error could not be calculated.

Evaluation of the historical campaign data resulted in the modification of normal operating procedures for ISE Campaign 1. The major changes were:

1. An attempt was made to improve characterization of the feed materials.
2. All previously unmeasured (estimated) or difficult-to-measure streams were characterized by nondestructive assay methods.
3. Sufficient data and records were maintained to calculate adequate limits of error for each material stream.
4. The campaign was conducted on a clean-to-clean basis, i.e., the process line was cleaned both before and after the campaign.

B. ISE CAMPAIGN 1

1. General

The ISE Campaign 1 was directly associated with the fabrication of 28 fuel rods for the Edison Electrical Institute – General Electric Company Program for the Utilization of Plutonium in Boiling Water Reactors – Phase II. Four master blend lots of two plutonium enrichments (3.54 and 3.42% plutonium-to-oxide ratio) totaling 136 kg of mixed (uranium-plutonium) oxide were processed during the fuel fabrication.

2. Material Balance Calculations

Three different types of material balances were constructed for ISE Campaign 1. The first type, a trial material balance, was representative of the balances prepared during the historical campaigns, except only material streams measured by analytical chemistry and net weights were considered. The second balance type, NDA Balance 1, was based on a combination of net weights and analytical chemistry for feed, product, and some inventory streams. In addition, nondestructive measurements were used to define inadequately measured material streams. The third balance, NDA Balance 2, defined each material stream with the best available measurement. The limits of error associated with MUF (LEMUF) for each balance were obtained by propagating the random components of error for each measurement.

TABLE 4. ISE Plutonium Material Balance Summary

Fabrication Identity	Trial Balance			NDA Balance 1			NDA Balance 2		
	MUF		LEMUF ^(a)	MUF		LEMUF ^(a)	MUF		LEMUF ^(a)
	(%)	(gm Pu)	(gm Pu)	(%)	(gm Pu)	(gm Pu)	(%)	(gm Pu)	(gm Pu)
HISTORICAL									
Campaign A ^(b)	1.54	352.8	—	—	—	—	—	—	—
Campaign B ^(b)	0.46	7.0	—	—	—	—	—	—	—
ISE CAMPAIGN 1									
Enrichment 1	1.40	36.0	10.8	0.89	22.8	19.6	1.14	29.6	98.8
Enrichment 2	1.74	45.3	7.2	1.41	36.9	19.5	1.62	42.2	107.6
ISE CAMPAIGN 2									
Enrichment 1	0.67	11.0	5.9	0.24	4.0	5.6	—	—	—
Enrichment 2	0.67	8.0	6.7	0.10	1.2	6.6	—	—	—

(a) LEMUF AT 95% confidence.

(b) The balances contain some terms which were not measured.

3. Trial Material Balance

The trial material balances⁽²⁵⁾ for ISE Campaign 1 resulted in plutonium MUF values of 1.40 and 1.74% for enrichments 1 and 2, respectively. These MUF values were larger than the associated LEMUF (see Table 4 for details). Examination of the total oxide balances⁽²⁵⁾ indicated that the plutonium loss was much larger than the total oxide loss. As a result, the plutonium loss could not be explained by the loss of mixed (uranium-plutonium) oxide during processing. This information indicates that the plutonium MUF resulted from a plutonia loss or diversion, analytical bias in the plutonium measurement, or inaccurate input feed measurement. In view of the difficulty encountered with the feed measurements,⁽²⁶⁾ the latter appeared to be the most plausible explanation. Some portion of the MUF values was attributed to the waste stream which was only partially measured by net weights and analytical chemistry.

The ISE Campaign 1 plutonium MUF values (1.40 and 1.74% for enrichments 1 and 2, respectively) and the value for Historical Campaign A (1.54%) were of the same order of magnitude. This indicated that the trial material balances were a good example of the status of material balance accounting before ISE.

4. NDA Balance 1

The NDA Balance 1 for ISE Campaign 1 resulted in plutonium MUF values⁽²⁷⁾ of 0.89 and 1.41% for enrichments 1 and 2, respectively. The MUF values were larger than the associated LEMUF quantities (Table 4). As previously stated in the discussion of trial material balances, a plutonium MUF value larger than the associated LEMUF quantity is a result of a plutonium loss or diversion, analytical bias in the plutonium measurement, or an inaccurate measurement of material. The moisture adsorption problems associated with the plutonia feed indicate an inaccurate measurement of material to be the most likely cause.

Although the plutonium MUF values for NDA Balance 1 were larger than the associated LEMUF quantities, they were smaller than the values for the trial material balances (0.89 and 1.41 versus 1.40 and 1.74% for enrichments 1 and 2, respectively). A reduction in the plutonium MUF values occurred because measured values were used in place of previously unmeasured waste and inaccurately measured scrap material.

The LEMUF quantities on plutonium for NDA Balance 1 were larger than those obtained in the trial material balances (19.6 and 19.5 versus 10.8 and 7.2 grams, respectively) because the passive neutron assay data and their uncertainties⁽²⁸⁾ were incorporated into the balance.

5. NDA Balance 2

The NDA Balance 2 was prepared by replacing the plutonia feed measurements based on net weights and analytical chemistry results in NDA Balance 1 by calorimetry measurements. The balance resulted in plutonium MUF values⁽²⁹⁾ of 1.14 and 1.62% for enrichments 1 and 2, respectively. The associated LEMUF quantities were a factor of 2 to 3 larger than the MUF values. The large LEMUF quantities result from the large uncertainty on the calorimetry measurements of the plutonia feed (uncertainties in the Pu-238 and Am-241 isotopic determinations).

Even though the plutonium MUF values were smaller than the limits of error, the MUF values were larger than those obtained in NDA Balance 1 (1.14 and 1.62 versus 0.89 and 1.41% for enrichments 1 and 2, respectively). The increased MUF values were the result of slightly larger feed values obtained from the calorimetry measurements. Since adequate standards were not available and the Pu-238 and Am-241 analysis were orders of magnitude larger in uncertainty than desired, the plutonium calorimetry measurements may contain a bias, (see Section III for details).

6. Operating Modifications

The information and experience gained during ISE Campaign 1 resulted in modification of the ISE Campaign 1 procedures. The modifications for ISE Campaign 2 were:

1. The plutonia was equilibrated in the glove box atmosphere to provide a uniform moisture content.
2. The method of determining the weighing errors for the balances were modified to account for zero deflection as well as typical tare and processing weights.

3. A majority of the existing processing forms were redesigned and new forms developed to satisfy the requirements for the ISE material balance and LEMUF calculations.
4. The method of standardizing the passive neutron counting data was refined to correct for instrument stability.
5. Inclusion of a special nest design for comparison of analytical chemistry with passive neutron and gamma (sodium iodide) counting techniques for single pellets. In addition, the sources of error in the analytical chemistry measurements were included.

C. ISE CAMPAIGN 2

1. General

Twenty fuel rods for the Edison Electrical Institute — General Electric Company Program for Utilization of Plutonium in Boiling Water Reactors — Phase II were completed during this campaign. The 107 kg of mixed (uranium-plutonium) oxide consisted of two master blend lots each of a different enrichment (2.58 and 2.38% plutonium-to-oxide ratio).

2. Material Balance Calculation

Trial material balance and NDA Balance 1 calculations prepared for ISE Campaign 1 were also completed for ISE Campaign 2. The NDA Balance 2 was not made for this campaign because NDA Balance 1 was made with the best available measurements for each material stream.

3. Trial Material Balance

The trial material balances⁽³⁰⁾ for ISE Campaign 2 resulted in plutonium MUF values of 0.67 and 0.67% for enrichments 1 and 2, respectively. The MUF values were again as observed in ISE Campaign 1 larger than the associated LEMUF quantities, see Table 4. The most plausible explanation of the significant MUF values⁽³¹⁾ was that some of the waste and scrap were considered as part of MUF because they either were not measured or were inadequately measured by a combination of analytical chemistry and net weight values.

Comparison of the plutonium MUF values of ISE Campaign 2 (0.67 and 0.67% for enrichments 1 and 2, respectively) with those from ISE Campaign 1 (1.40 and 1.74% for enrichments 1 and 2, respectively) and Historical Campaign A (1.54%) indicated the ISE Campaign 2 values were a factor of 2 or 3 smaller even though the throughputs of each enrichment in ISE Campaign 1 (~ 2.6 kg Pu) and Historical Campaign A (~ 23.0 kg Pu) were larger than each enrichment in ISE Campaign 2 (~ 1.3 kg Pu). The major reduction in plutonium MUF values was attributed to the equilibration of the plutonia feed in ISE Campaign 2 which eliminated the measurement bias caused by moisture adsorption. The remaining plutonium MUF reduction was attributed to the differences in throughput rates, improved measurement capability, and "chance" variations.

4. NDA Balance 1

The NDA Balance 1⁽³²⁾ for ISE Campaign 2 resulted in plutonium MUF values of 0.24 and 0.10% for enrichments 1 and 2, respectively. Since the range on the LEMUF quantities included zero, no significant loss of material could be detected because of the measurement uncertainties in the system.

The plutonium MUF values for NDA Balance 1 (0.24 and 0.10% for enrichments 1 and 2, respectively) were somewhat smaller than those in the trial balance (0.67 and 0.67% for enrichments 1 and 2, respectively). The reduction in plutonium MUF values was a result of the incorporation of nondestructive assay measurements for previously unmeasured or inadequately measured heterogeneous streams.

A comparison of the NDA balances in ISE Campaigns 1 and 2 indicated that ISE Campaign 2 plutonium MUF values were 4 to 16 times smaller than those in ISE Campaign 1. The major reduction in the plutonium MUF values appeared to result from the elimination of the bias in plutonia feed measurement. The remaining plutonium MUF reduction resulted from improved measurements or by "by chance" variations. These comparisons between ISE Campaigns must be made with the knowledge that the plutonium throughput rates in each campaign were different by a factor of 2 (~ 2.6 kg plutonium per enrichment in ISE Campaign 1 and ~ 1.3 kg plutonium per enrichment in ISE Campaign 2).

V. SPECIAL TESTS

A. ISE-2 NESTED DESIGN

Since the ISE was undertaken to evaluate the effectiveness of safeguard control practices based on measured material balances, the measurement uncertainty in the analysis for plutonium content in mixed (uranium-plutonium) oxide fuel was an important safeguard consideration. This uncertainty was the major contributor to the limits of error associated with each material stream utilized in material balance calculations. Since a considerable amount of analytical work was required in defining these material streams, an estimate of the source and magnitude of the measurement error was required to complete the analysis.

A statistical testing procedure known as a "nested design" was used to estimate random components of error in the measurement methods used to establish the material balances.⁽³³⁾ In addition, the nested design technique provided an opportunity to compare data obtained by several nondestructive measurement methods with data from the controlled potential coulometer method as well as the determination of the sources of error in the coulometric analysis method.

The nested design consisted of 18 whole pellets which were chemically analyzed by controlled potential coulometry as part of the fuel fabrication quality control program (process pellets) and 18 pellets which were given special homogenizing treatments (experimental pellets). The 18 experimental pellets were first encapsulated and measured in random order by nondestructive gamma and neutron counting methods. These same pellets were then chemically dissolved and measured by the controlled potential coulometric method.

The methods studied can be ranked with respect to random components of measurement error for a single Pu/MO₂ determination by dividing the methods into four groups of about equal precision.

Most precise: (0.3% relative)	— Controlled potential coulometry.
Next most: (1.0% relative)	— Nondestructive gamma counting at 0.414 MeV for whole pellets. — Nondestructive gamma counting for the sum of the 0.414- and 0.208-MeV counts from whole pellets. — Nondestructive gamma counting at 0.208 for whole pellets.
Less precise: (1.5% relative)	— Nondestructive gross neutron counting with any pellet preparation.
Least precise: (0.8 to 3% relative)	— Nondestructive gamma counting at 0.414 MeV for both halved and pulverized pellets. — Nondestructive gamma counting for the sum of the 0.414 and 0.208 MeV counts from halved and pulverized pellets. — Nondestructive coincident neutron counting for any pellet preparation. — Nondestructive gamma counting at 0.208 MeV for halved and pulverized pellets.

The major sources of measurement error in the controlled potential coulometric method were divided equally between dissolution errors and aliquanting errors (including coulometer error with a small contribution from sampling error).

B. MECHANIZED ACCOUNTING AND CRITICALITY SYSTEM

The term "mechanized accounting" is frequently applied to a computerized inventory control system for nuclear materials. Late in the ISE, a mechanized accounting system was brought on line. While the results were not used for the material balance calculations, the feasibility and usefulness of mechanized accounting were demonstrated. In addition, many of the control concepts utilized in material balance calculations were built into the machine programs.^(34,35) The mechanized accounting system is now in routine use.

One of the major benefits realized from mechanized accounting was a detailed understanding of the essential elements of material control. This included sampling points, sampling frequency, measurement capability, and the requirements for completion of measured material balances. In addition, the routine propagation of errors for all components of the material balance equation served as a sensitivity analysis for identification of items of major importance. The ability of the computer to sort data was used to prepare reports which allowed manual study of inventory control functions.

The computer program also provided the capability to build logical controls into the system. For example, the flow of material can be traced from start to finish. If this route is broken and restarted from another point, the error will be identified to the control operator. In addition, there are a large number of computer cross checks which are not possible with manual methods. Thus, consistency within the data is a useful control criteria.

The experience with the existing mechanized accounting system has indicated the potential benefits to be obtained from the use of heuristic control limits by work station. This type of control may provide timely and sensitive warning of unusual material loss.

The control of a dynamic mixed (uranium-plutonium) oxide inventory based on measurements is complicated by a wide variety of "special cases." These cases result in a complex program for material control with many alternate paths which are selected on the basis of previous processing history. In addition, the control procedures necessary to guarantee the calibration of a variety of measurements, while not unique to mechanized accounting, is intensified by the number of measurements which are handled by the mechanized system.

VI. PROGRAM EVALUATION

A. NONDESTRUCTIVE MEASUREMENTS

Promising nondestructive assay techniques were evaluated under in-plant fuel processing conditions during this program. Thirty-two typical mixed (uranium-plutonium) oxide fuel rod, scrap, and waste-discard standards were prepared. The available measurement techniques and assay equipment which included calorimetry, passive neutron, and passive gamma methods, were qualified on fuel standards. The usefulness and acceptability of the assay techniques as a tool for material balance accounting were demonstrated during ISE Campaigns 1 and 2.

B. MATERIAL BALANCES

1. Historical Material Balances

The historical data and operating procedures for two campaigns were provided to the ISE — Technical Representatives. These data were examined to determine the status of material balance accounting at General Electric. Material balances calculated from the data were used as a base case for methods of calculations before the ISE.

2. ISE Campaign Material Balances

The material balances calculated during the ISE campaigns demonstrated it was possible to prepare measured material balances by determining the plutonium content of all identifiable material streams. To complete these balances, it was necessary to define the waste and some of the scrap streams by nondestructive assay values. The plutonium MUF values obtained for the balances were smaller than the associated limits of error obtained by the propagation of the random components of the measurement error. These balances demonstrated the possibility of obtaining MUF values of less than 0.5% by material balance accounting.

3. Unit Processing Operation Material Balances

One of the planned objectives of ISE Campaign 2 was to measure the plutonium losses during selected unit processing operations and to the degree possible, identify the loss mechanisms. The plutonium losses were to be determined by

measuring the changes in net weight and chemical composition resulting from the unit processing operation. Evaluation of ISE Campaign 1 data indicated the expected losses were too small to be identified because of measurement error and this objective was deleted from the Integrated Safeguards Experiment.

C. DATA SYSTEMS

The Mechanized Accountability and Criticality System was not fully operational during the ISE Campaigns. The system has been continuously updated to demonstrate inventory and material flow control. There is a strong indication this system may prove useful in instituting new material control and safeguards techniques.⁽³⁴⁾

D. ACCURACY AND PRECISION REPORT

Another objective of the program was to provide a nonproprietary report on the accuracy and precision of the measurements made during the ISE. This report was to include: (a) type of material, (b) kind of instrument, (c) measurement method, (d) calibration method and frequency, (e) calibration standards used, (f) computational procedures used on measurement and calibration data, (g) accuracy (bias) of each method, and (h) precision for each method.

Essentially all of the necessary information was collected and the precision values calculated during the preparation of the material balances. The responsibility for the final report was assigned to the ISE — Technical Representatives.

VII. PROGRAM IMPLICATIONS

A. PROGRAM LIMITATIONS

Although the material balances calculated during ISE Campaigns 1 and 2 were based on data obtained during routine processing, the results are difficult to extrapolate to a manufacturing scale. The sampling levels used for product certification of the demonstration fuel were much higher than those anticipated for the day-by-day operation of a plutonium fuel factory. As a result, the measurement errors associated with the analytical chemistry determination for plutonium are probably understated and result in smaller limits of error on MUF. The operating personnel associated with this program were attuned to development-type activities and constantly alert for unusual and extraordinary occurrences. This degree of surveillance is not expected from production personnel in a large-scale plutonium fuel factory.

In contrast to the possible understatement of the errors in the material balances, the errors for the nondestructive assay techniques are probably larger than those expected in a plutonium fuel factory. Many of the measurement techniques were new and the PIP demonstration equipment was not always optimized for the materials measured. As a result, the errors associated with each measurement could probably be reduced if the equipment was designed for a specific application. Although the nondestructive assay techniques were used to measure only a small percentage of the plutonium in the material balance, improvements in measurement capabilities should reduce the limits of error on future material balances.

The limits of error for the ISE Campaign 1 and 2 balances include only the random components of measurement error and do not include the systematic component of the total measurement error.⁽³⁵⁾ The systematic component of error must be included if material balances are to be used as a safeguards tool.

B. NONDESTRUCTIVE MEASUREMENTS

Each nondestructive assay technique has advantages and limitations which must be judged against the specific requirements of safeguards or process control applications or both. No single method can satisfy all requirements. The PIP and ISE have effectively demonstrated that available nuclear assay instrumentation can satisfy many of today's safeguards and process control requirements. With continued development, nondestructive systems should have greater capability and usefulness in the future. Certain systems appear ready for use by industry today, while others may

require additional development to attain the level of accuracy required by current safeguards requirements. In either case, it is important to note that all nondestructive measurements are relative to known standards. The degree of sophistication required for each nondestructive measurement depends on the level of detection and accuracy required. Nondestructive analyses costs are in general lower than those for analytical chemistry. On this basis, the nuclear fuels industry should more fully evaluate the feasibility of utilizing the available nondestructive assay capabilities for the routine measurement of plutonium-bearing material.

To integrate nondestructive assay techniques into routine use in the nuclear industry, it is necessary to establish uniform procedures and data reduction methods. A step toward the solution of these problems would be the preparation of well-characterized standards to provide a basis for evaluating nondestructive assay techniques. Only when nondestructive assay methods are utilized on a standardized basis can sufficient operating experience be generated to gain confidence and define more complex capabilities for future applications.

C. EVALUATION OF MATERIAL BALANCE TERMS

The first material balances prepared for ISE Campaigns 1 and 2 were trial balances which were based on the accounting practices utilized prior to the Integrated Safeguards Experiment. These balances were considered as reference cases and were used as a basis for evaluating improvements made during the program. A comparison of the trial balances with NDA Balances 1 and 2 indicated a reduction in the contribution to plutonium MUF values occurred during the material balance calculations (see Table 5).

TABLE 5. Effects of Measurements on MUF (Table entries in gram Pu)

	ISE Campaign 1		ISE Campaign 2	MUF Reductions	
	Actual	Normalized*	Actual	Actual	Normalized*
Trial Balance	81.3	41.6	19.0	62.3	22.6
NDA-1 Balance	59.7	30.6	5.2	54.5	25.4
MUF Reductions	21.6	11.0	13.8	—	—

* Adjusted to ISE Campaign 2 plutonium feed value of 2443.1 gram.

The ISE Campaign 1 data was normalized to the ISE Campaign 2 plutonium throughput data for comparison, with the knowledge that decreases in throughput rates are normally associated with decreases in absolute MUF values and increases in relative MUF values. A review of the two ISE Campaigns indicate that the relative uncertainties associated with the major terms in the material balances were of similar magnitude (feed — 0.02 versus 0.03% and product 0.35 versus 0.45% for ISE Campaigns 1 and 2, respectively). These similarities indicated that normalizing the balances to the same base does not result in gross differences.

A comparison of the normalized MUF values for ISE Campaign 1 with the MUF values for ISE Campaign 2 (comparison of rows in Table 5) indicated a reduction in the contribution to MUF of 22.6 and 25.4 grams plutonium for the trial and NDA-1 balances, respectively. Since the reduction of the contributions to MUF for both the trial balance and NDA-1 balance was on the magnitude of 20 grams plutonium, the cause of MUF reduction was probably similar for each balance. A major portion of the reduction in contributions to MUF for ISE Campaign 2 was attributed to the elimination of a bias in the plutonia feed measurement by equilibration for moisture content prior to sampling. Calculations based on the moisture pickup during plutonia equilibration for this Campaign indicated the bias should be on the order of 15 grams of plutonium. The remaining 5 to 10 grams of plutonium which comprises the reduction in the contribution to MUF was attributed to improved handling and measurement accuracy during ISE Campaign 2.

A comparison of trial balances and NDA-1 balances within campaigns (comparison of columns in Table 5, normalized columns for ISE Campaign 1) indicated a reduction in the contributions to MUF of 11.0 and 13.8 grams of plutonium for ISE Campaigns 1 and 2, respectively. The reductions resulted from shifting the expected value of MUF from some positive value to zero for NDA Balance 1 by measuring the heterogeneous streams (dirty scrap and waste-discards) by NDA techniques instead of the estimation techniques used in the reference trial balances.

A fuel fabricator could calculate a material balance on a difference basis with only measurement of feed, product, and inventory. When material balance terms are arrived at on a by difference (waste, in this case), they must be classified as MUF with zero uncertainty for the limit of error. Assuming no significant measurement bias, the MUF value will most likely be larger than the associated limit of error. This condition is unacceptable, if the MUF value is not trivial, because it signals a potential material processing loss or diversion. The measurement of material in the by-difference category changes its classification from MUF to "material accounted for" resulting in a reduction of the MUF value. Since the measurement uncertainty on this material is now greater than zero, the limit of error on MUF is increased.

The propagation of all measurement errors should result in a LEMUF quantity which contains the MUF value. If this is not the case, the material balance indicates one or more of the following may have occurred:

1. processing material loss,
2. material diversion,
3. measurement bias.

If a fuel fabricator utilizes measured material balances as a technique to detect material loss or diversion, he must first assign primary priority to measurement of all major material streams to reduce MUF as much as possible. Further reductions in MUF with associated increases in the LEMUF quantity can be obtained by measuring waste material. These relationships provide the incentive for the fuel fabricator to adequately measure the major streams before placing major emphasis on the measurement of waste streams.

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PREVIOUS REPORTS IN THIS SERIES

1. Plant Instrumentation Program First Quarterly Report, April 1970 to June 1970, GEAP-12114-1.
2. Integrated Safeguards Experiment First Quarterly Report, May 1970 to August 1970, GEAP-12114-2.
3. Plant Instrumentation Program Second Quarterly Report, July 1970 to September 1970, GEAP-12114-3.
4. Integrated Safeguards Experiment Second Quarterly Report, September–November 1970, GEAP-12114-4.
5. Plant Instrumentation Program Third Quarterly Report, October–December 1970, GEAP-12114-5.
6. Integrated Safeguards Experiment Third Quarterly Report, December 1970 – March 1971, GEAP-12114-6.
7. Integrated Safeguards Experiment Fourth Quarterly Report, April–June 1970, July 1971, GEAP-12114-7.

REFERENCES

1. GEAP-12114-3, Table B-1, page 26.
2. GEAP-12114-7, page 7.
3. GEAP-12114-3, page 10.
4. GEAP-12114-7, page 8.
5. Sher, R., "Operating Characteristics of Neutron Well Coincident Counters," Brookhaven National Laboratory, BNL-50332 (January 1972).
6. Omohundro, R. J, and Marchetti, F. A., "A Prototype Coincidence Neutron Counter for International Safeguards and Arms Control Application," NRL Memorandum Report 2005, dated June 1969.
7. Strain, C. V., and Omohundro, R. J., "Coincident Neutron Equipment," NRL Memorandum Report 2107, dated March 1970.
8. WCAP-7562-4 Plant Instrumentation Program. Final Quarterly Report, March 3, 1971.
9. GEAP-12114-3, pages 13-16.
10. GEAP-12114-1, 3, 5, 6, and 7.
11. GEAP-12114-7, page 8.
12. Higinbotham, W., "Passive Gamma Ray Scanning of Plutonium Recycle Fuel Rods," Brookhaven National Laboratory, BNL-50351 (June 1972).
13. GEAP-12114-4, page 9.
14. GEAP-12114-7, page 10.
15. GEAP-12114-1, page 4.
16. GEAP-12114-3, page 14.
17. GEAP-12114-7, page 6.
18. GEAP-12114-3, pages 13-16.
19. GEAP-12114-1, page 4.
20. GEAP-12114-6, Appendix A, page 10.
21. GEAP-12114-3, Table B-3.
22. GEAP-12114-6, page 3.
23. GEAP-12114-4, page 4.
24. *Ibid.*, page 3.
25. *Ibid.*, pages 10-12.

26. *Ibid.*, pages 8—10.
27. GEAP-12114-7, pages 9—11.
28. *Ibid.*, pages 2—6.
29. *Ibid.*, pages 10 and 12—13.
30. *Ibid.*, pages 13—16.
31. *Ibid.*, pages 15—16.
32. *Ibid.*, pages 14 and 17.
33. Hagie, L. T., Bishop, D. M., Nelson, W. B., Robles, M. N., and Wadekamper, D. C., "Measurement Error in Several Pu Assay Methods Based on a Balanced Staggered Nested Design," GEAP-12283, March 1972.
34. Hagie, L. T., "An Operating Computerized Safeguards and Criticality System for Plutonia and Urania," *Transactions of the American Nuclear Society*, 13, No. 2, p 750 (Nov. 1970).
35. Smith, C. N., "Limit of Error Calculations Used in Integrated Safeguards Experiment," Transactions of the Institute of Nuclear Materials Management Twelfth Annual Meeting, Vol. 1, June 29—July 1, 1971, pp 193—207.
36. Hagie, L. T., Wadekamper, D. C., and Marcuse, W., "Calculated Inventory Difference — A Potential Safeguards and Material Control Tool," Transactions of the Institute of Nuclear Materials Management Twelfth Annual Meeting, Vol. 1, June 29—July 1, 1971, pp 270—301.

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