

# NATIONAL BUREAU OF STANDARDS REPORT

10 872

## INTEGRATED SAFEGUARDS EXPERIMENT

Campaign 2 Report



U.S. DEPARTMENT OF COMMERCE  
NATIONAL BUREAU OF STANDARDS

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# NATIONAL BUREAU OF STANDARDS REPORT

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NBS REPORT

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## INTEGRATED SAFEGUARDS EXPERIMENT

Campaign 2 Report

Frederick J. Perella  
Charles N. Smith

Approved:

*W. H. Hight*  
Technical Analysis Division

Approved:

*Richard T. Lange*  
Technical Analysis Division

Approved:

*Charles N. Smith*  
Technical Analysis Division

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U.S. DEPARTMENT OF COMMERCE

NATIONAL BUREAU OF STANDARDS



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## ABSTRACT

The Integrated Safeguards Experiment (ISE), sponsored by the USAEC (Division of Nuclear Material Security) was performed at the General Electric Company, Vallecitos Nuclear Center, (GE-VNC) Pleasanton, California.

It was a collaborative project with General Electric Company (GE-VNC), Brookhaven National Laboratory, Technical Support Organization (TSO-BNL) and the National Bureau of Standards, Technical Analysis Division (NBS-TAD) being the principle participants. Physical measurements were made and analyzed by GE-VNC. The experimental plan used in the experiment was the responsibility of TSO-BNL and NBS-TAD. Experimental support of the non-destructive analysis (NDA) equipment was provided by TSO and technical and analytical guidance was provided by NBS and TSO.

The ISE was conducted in two separate campaigns, designated Campaigns 1 and 2. Each Campaign, carried out in a production environment, involved the fabrication of mixed oxide  $\text{PuO}_2\text{-UO}_2$  fuel rods scheduled for use in a public utility power plant reactor. The purpose of ISE was to evaluate the usefulness to Safeguards of a material balance based on completely measured values for materials containing plutonium.

The material balance for Campaign 1 was calculated and the result reported in NBS report 10 660. Also the impact of the use of non-destructive assay equipment and methods, which made possible this measured material balance, was studied and evaluated.

Campaign 2 procedures followed closely those of Campaign 1, differing only in that certain procedures were improved as suggested by Campaign 1 experience. These changes, and their effect on the material balance, are reported. The significant improvement over the results obtained for Campaign 1 are noted and reported.

In addition, Campaign 2 included a nested design experiment which had as its purpose to identify and determine the magnitude of the several variance components which contributed to the total measurement error when determining plutonium content of  $\text{PuO}_2 - \text{UO}_2$  pellets by the controlled potential coulometric method (wet chemistry). Prior to destructive analysis, these same pellets were measured by non-destructive gamma and neutron counting methods. A comparison of results of wet chemistry (destructive) and non-destructive measurements is reported.



## Overview

The nuclear materials safeguards program to guard against the loss or unauthorized diversion of nuclear materials has long been hampered by the lack of material balances based entirely on quantitative measurements. The results of the Technical Studies program in the development of non-destructive assay instruments have made possible the timely measurement of many hard-to-measure materials, leading to the possibility of completely measured material balances.

The Integrated Safeguards Experiment (ISE) was established at the General Electric (GE) Vallecitos Nuclear Center (VNC) with the broad purpose of testing the usefulness, to Safeguards, of materials balance accounting techniques which employ data based completely on quantitative measurement. The concept is that, if all streams and outlets of fissile product are accurately determined and the uncertainties associated with each measurement accounted for, then such material balances would provide control over nuclear materials within a production facility with a known precision. If such balances can be made often enough, and if the balances are precise enough, then to what extent might this satisfy

the requirements of the Domestic Safeguards Program and specifically for plutonium fuel processing plants?

The ISE was planned to be run in several campaigns, with changes and refinements in procedures to be added to each succeeding campaign as suggested by experience in the previous one. Campaign 1 followed a plan carefully developed by GE and approved by the AEC and other participating ISE representatives. In addition, the Integrated Safeguards Experiment team followed details of the work very closely and thus were able to detect problems with the procedures, as well as error sources which would require corrective action to insure more accurate material balances.\* Campaign 2 of the Integrated Safeguards Experiment was tentatively planned to begin several weeks after completion of Campaign 1, allowing ample time to analyze Campaign 1 data and to introduce changes in procedures as suggested by the results of Campaign 1 experience.

Since fuel rods fabricated during the ISE were to be produced for a General Electric Company customer in accordance with an agreed upon schedule, and since chemical laboratory tests and analysis for Campaign 1 consumed considerably more time than anticipated, Campaign 2 processing was begun before complete results of Campaign 1 were available.

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For detailed discussion and results of Campaign 1, refer to NBS Report number 10 660, January 1972.

This prevented the preparation of a formal experimental plan to be submitted to the ISE representatives\* for approval before Campaign 2 was begun.

The proposed experimental plan for Campaign 2 was therefore informally discussed and items for inclusion verbally agreed to. Because of this pre-campaign planning lacked sufficient depth, resulting in several areas being neglected. For example, insufficient data, primarily in the non-destructive testing area, precluded a proper evaluation of test procedures, hence there were no changes introduced in the Campaign 2 NDA of scrap and waste materials. This was reflected in the results, the uncertainties associated with Campaign 2 scrap and waste values being essentially the same and, in some cases, even larger than those of Campaign 1.

The Campaign 2 plan covered all features of the Campaign 1 experimental plan and, in addition, included features which experience in Campaign 1 indicated were necessary and essential for more accurate material control.

These additional features were as follows:

1. Re-organized, more detailed data forms for recording measurements, calibration data, and material transfers to and from all areas within the plant.

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\*  
AEC, TSO and NBS members assigned to the ISE.

The objective was to reduce to a minimum, errors caused by incomplete and faulty recording of data, which was an error source in Campaign 1.

2. A program for multiple measurements including both NDA and wet chemistry, of plutonium feed. Campaign 1 experience indicated that it was virtually impossible to produce a meaningful material balance if the plutonium feed material was not prepared and measured in this manner.

3. A replicate sample - replicate analysis program for sintered pellets (nested design experiment). This was found necessary in order to identify, segregate and determine the magnitude of the error sources when measuring Pu content of sintered pellets by several methods.

4. An exhaust filter measurement program.

5. A reorganized, more detailed and more stringent calibration program. This included more frequent calibration exercises as proposed by ISE representatives.

Campaign 2 involved the processing of approximately 100 kilograms of mixed oxide fuel. The oxide contained 80 percent fissile plutonium and was comprised of two enrichments\*, 2.58 and 2.38 percent, each consisting of approximately 50 kilograms.

Since results of Campaign 1 indicated that the major error sources were due to deficiencies in the measurement field, special emphasis in Campaign 2 was put on means of reducing these errors. The multiple measurement program

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Enrichment defined as  $\text{Pu}/(\text{U}+\text{Pu})$



for feed, which involved many individual measurements, resulted in much better agreement between these measurements than was observed for those in Campaign 1. This was attributed almost entirely to the equilibration treatment which simply subjected the plutonium dioxide powder to glove box atmosphere until it adjusted to the relative humidity of the box before sampling. The agreement between GE-VNC and New Brunswick Laboratories for wet chemistry assay values of the same sample material did not, however, show any appreciable improvement, which suggests a need for standardization of measurement methods and sampling. The NDT nested design experiment (replicate sample-replicate analysis) was accomplished with pointed results. From information obtained during Campaign 1 it was not discernible whether the observed variation between samples within a batch was due to variation within a measurement method or if the batch was actually inhomogeneous. This experiment characterized the measurement error in gamma and neutron counting techniques as well as for the controlled potential coulometric method. Total percent coefficient of variation of 0.28 reported by G.E. was obtained for the controlled potential coulometric method for  $\text{Pu}/(\text{UO}_2 + \text{PuO}_2)$  and a small bias was observed between the process and experimental sample results, both measured by the coulometric method. This bias reflects differences between analysis periods. Within-pellet variation and between-boat variation



were found to be insignificant. Between-pellet variation was as large as the dissolution variance. The analytical variation was approximately half that of the dissolution variance. A substantial difference between the total percent CV for processed pellets vs nested design measurements may be due to more careful handling during the experiment.

Of the two passive counting methods investigated, gamma counting with NaI(T) detectors had a smaller uncertainty than neutron counting, but gamma counting was more sensitive to the sample configuration.

The material balance, as shown in this report, was calculated by the NBS representative and care was taken to include measurements of all streams except those taken on the absolute filters. These measurements were considered unreliable and the plutonium content of these filters was therefore considered as MUF. The material balance calculated for the total Campaign, indicated a MUF of 9.3 grams plutonium with the limit of error of 14.4 grams Plutonium. LEMUF represents 0.6 percent of throughput (Pu feed). Since LEMUF for Campaign 1 amounted to 4.0 percent of throughput it is readily seen that the degree of material control achieved during Campaign 2 was significantly better than that of Campaign 1.

## REPORT ON CAMPAIGN 2 OF THE INTEGRATED SAFEGUARDS EXPERIMENT

### I. Introduction

#### A. Background and Objectives

The Integrated Safeguards Experiment (ISE) was conducted at the General Electric Company, Vallecitos Nuclear Center, (GE-VNC) Pleasanton, California, during the period August through December 1970. The purpose of the ISE was to test the usefulness, to Safeguards, of material balance accounting techniques which employ data based completely on quantitative measurements. The ISE was planned to accommodate four major objectives:

1. Evaluate the impact of recently developed non-destructive assay methods on material balance accounting.
2. Study methods by which measurement data are acquired, processed and analyzed in the facility's nuclear material control system.
3. Evaluate the accuracy and timeliness of currently employed chemical analysis and sampling.
4. Determine the magnitude of unidentified process losses for specific unit processes and possible methods for reduction and isolation of these losses through use of submaterial balance areas.

A secondary objective was to determine the possibility of utilizing the results and experience from an ISE to formu-

late numerical criteria and standard procedures for an "across the board" use in AEC licensee plants.

The main effort to gain these objectives was applied during Campaign 1 of this experiment. The broad objectives set forth for the ISE were essentially gained during Campaign 1. Findings and conclusions on those objectives were reported in NBS report 10 660. This report also points out that objective No. 4, "to determine the magnitude of unidentified process losses for specific unit processes and possible methods of reducing them" was not realized due to the high uncertainties in the  $\text{PuO}_2$  feed measurements thus causing unrealistic LEMUF values. This problem was corrected prior to start-up of Campaign 2. However, because of limitations in time and funds, the investigation of unit process losses was dropped from Campaign 2 procedures. The planning, project description, detailed procedures and background features of the ISE, as well as the results of Campaign 1, are reported in NBS Report 10 660.

Campaign 2 procedures followed closely those of Campaign 1 and therefore will not be discussed in detail in this report. The primary objective of Campaign 2 therefore, was to repeat Campaign 1 procedures, incorporating changes, refinements and other helpful features as suggested by the results of Campaign 1, and to compare the results of both campaigns.

## B. Description of Plant and Process Operations

The Plutonium Fuel Laboratory is comprised of two sections as indicated on the floor plan, Figure 1. The equipment on the right half is primarily for fabrication of limited quantities of mixed oxide ceramic fuel for specific test purposes. In this area also are included facilities for material and property studies and auxiliary fuel fabrication operations. The equipment in the left half of the laboratory is primarily for fabrication of test and demonstration fuel for both fast and thermal reactors. It is in this half of the laboratory that the Integrated Safeguards Experiment was conducted. The work was done exclusively in a self-contained, interconnected glove box line. Each individual box contains two inlet and two outlet ports, each being covered with an absolute filter. In addition, the outlet port absolute filters are preceded by double thickness prefilters. Material flow and process operation in individual glove boxes are shown in Figure 2.

## C. Measurements

Measurement effort for Campaign 2 was considerably more extensive and more carefully carried out than that of Campaign 1. These measurements included wet chemistry for both plutonium and uranium and non-destructive testing, utilizing calorimetry, neutron counting and gamma counting. A summary of the number and type of measurements made on each stream is shown in Table 1. Also, points in the process line, material involved,



and types of measurements made are shown in Figure 3.

It should be noted that chemical assay values for Pu content of finished rods, sintered recycle material and clean scrap were assumed to be the same as for the production material.



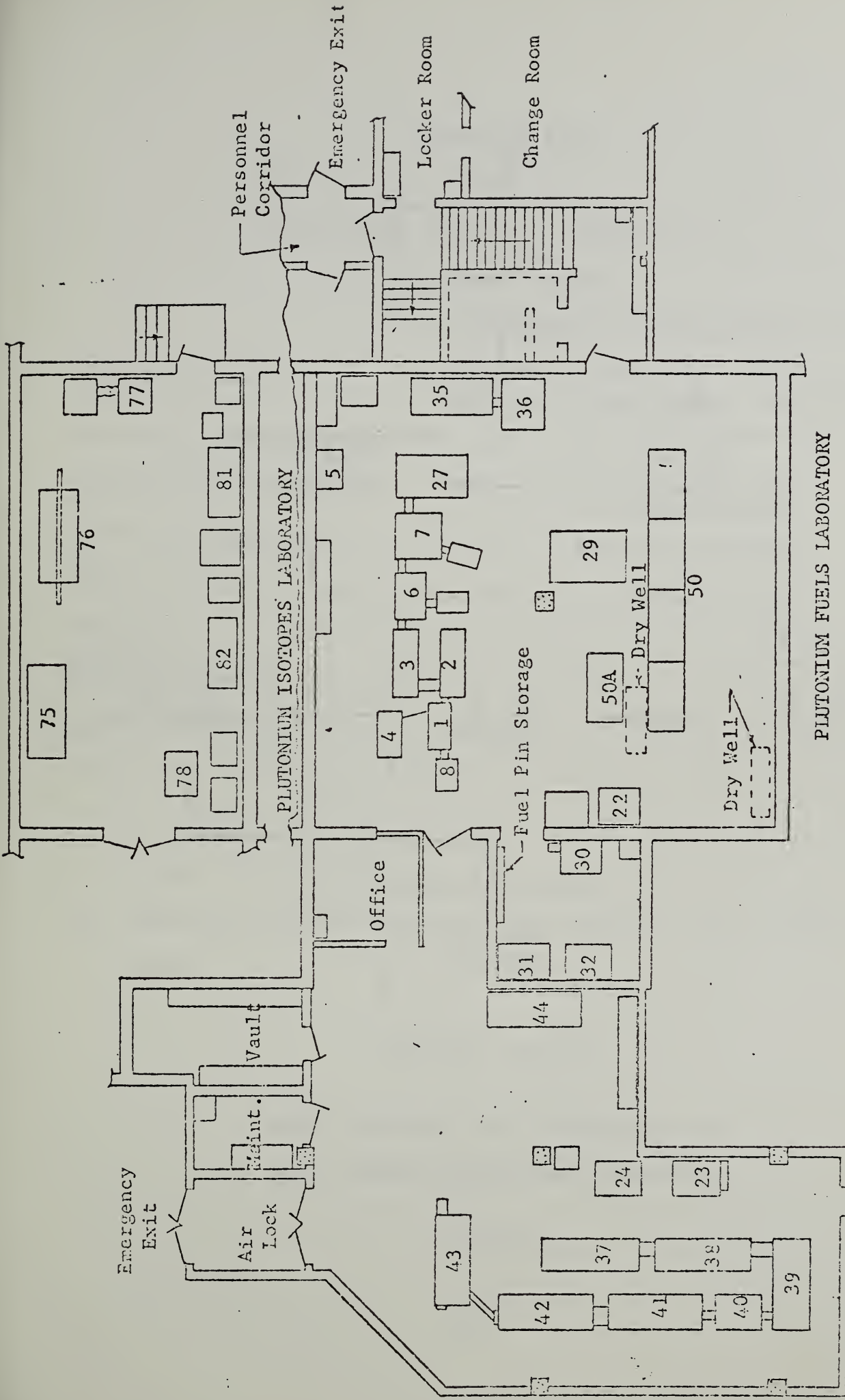
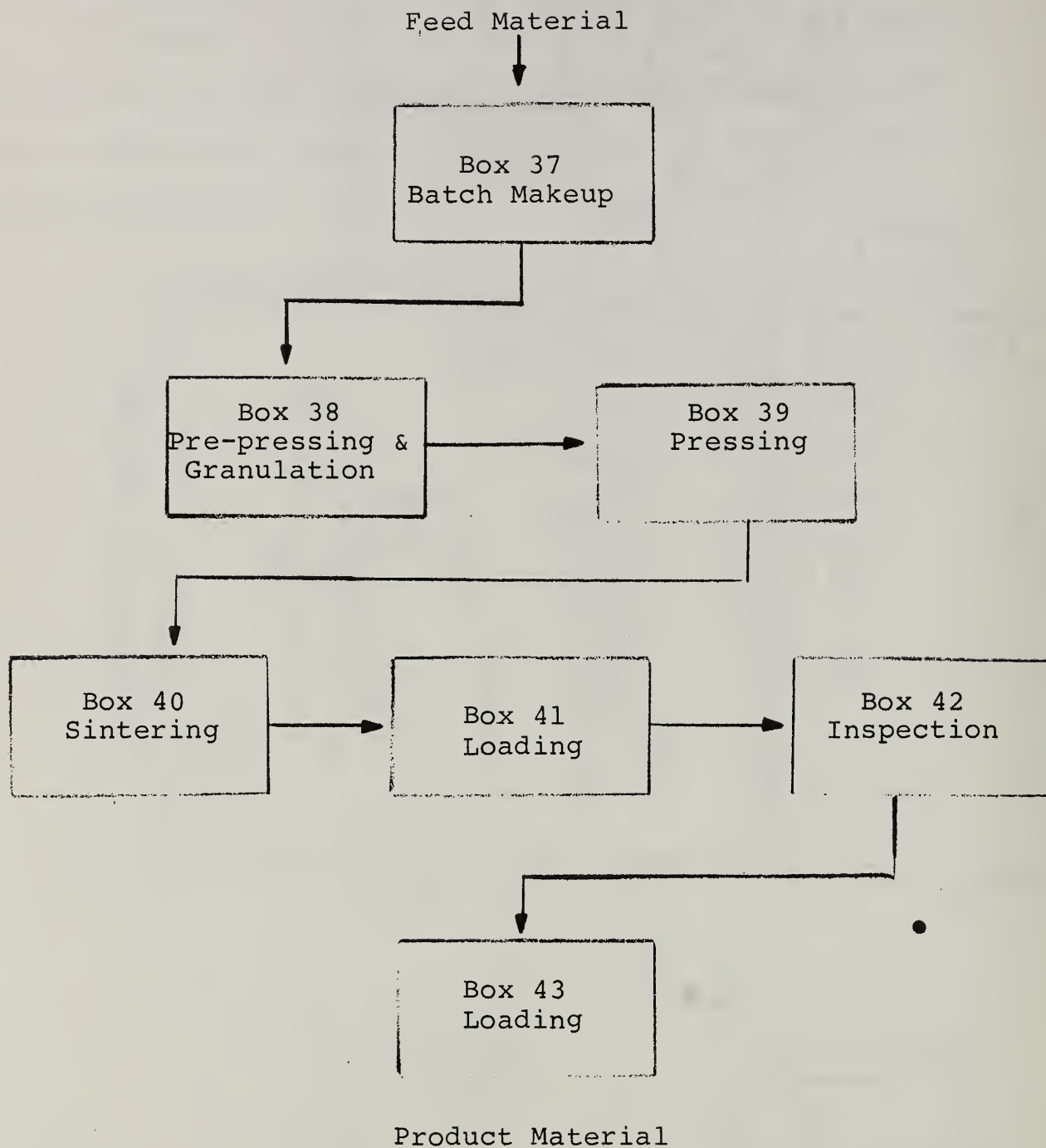


Figure 1: GE-VNC Plutonium Fuel Laboratory Floor Plan



Arrows denote main material flow  
Plutonium Fuels Fabrication Line

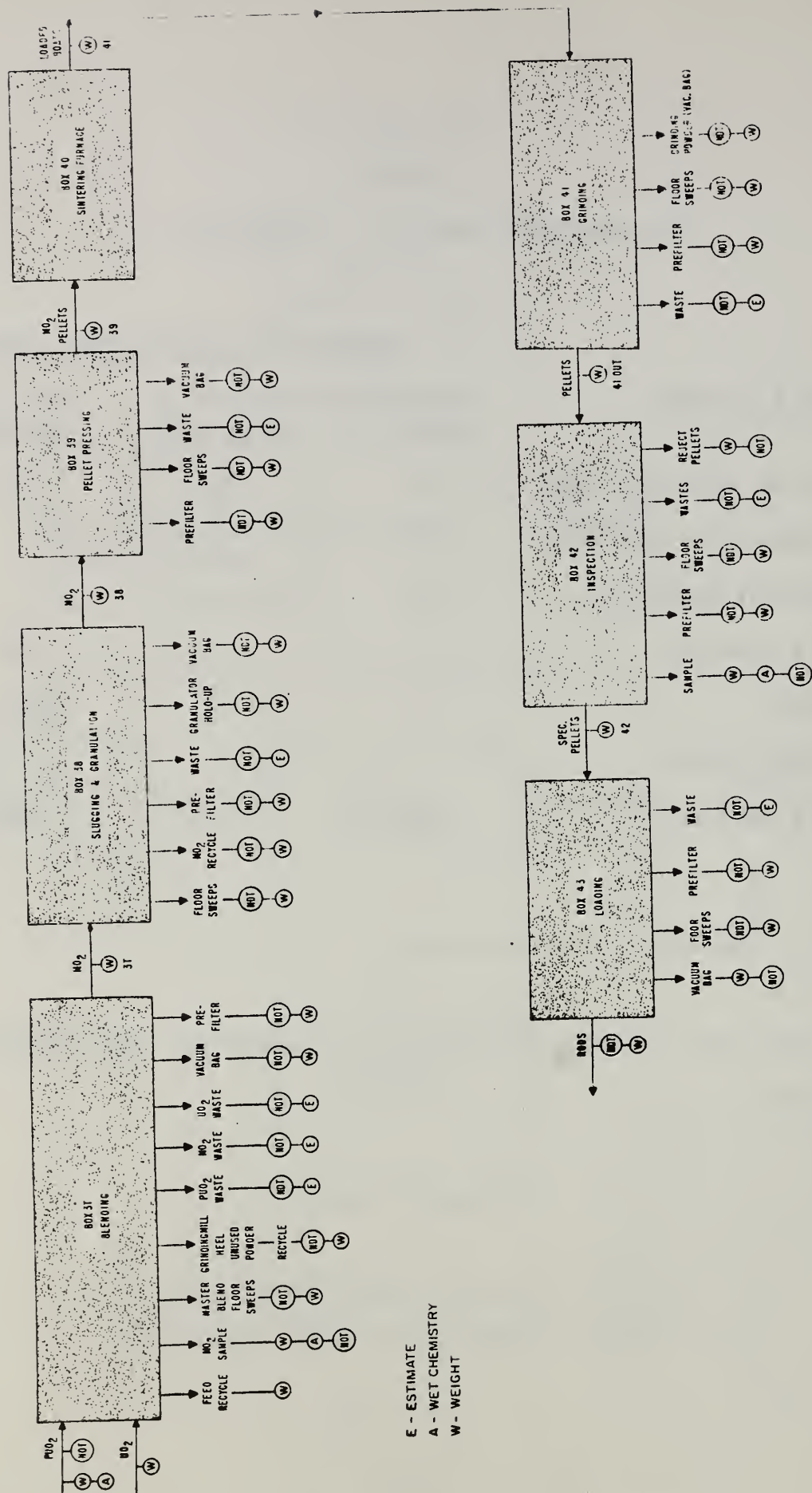
Figure 2

Table 1

## Measurement Summary - Campaign 2

<u>Material Stream</u>	<u>Number of measurement determinations</u> <sup>(a)</sup>			
	<u>Chemical Assay</u>	<u>calorimetry</u>	<u>neutron counting</u>	<u>gamma counting</u>
Plutonium Dioxide feed	16	11	35	-
Preproduction Samples	17	-	-	-
Production Samples	33	-	-	-
Scrap & Recycle	-	-	48	-
Discards	-	-	104	-
Completed Rods	-	-	-	20
Special Samples	36	10	46	72

(a) Does not include calibration, standard, and background runs.



E - ESTIMATE  
A - WET CHEMISTRY  
W - WEIGHT

Figure 3  
Points and Types of Measurements

## II. Material Balance - Discussion of Inputs

### A. Feed Material Measurements

The plutonium feed ( $\text{PuO}_2$ ) was contained in three sealed cans. This material was assayed, as received, by the calorimetry and neutron counting methods. Calorimetry measurements pointed up a large isotopic discrepancy which was resolved with the vendor before the cans were opened to the process line. This feature demonstrated the importance and usefulness of calorimetry in resolving shipper-receiver differences on undisturbed feed material. The cans were then opened, contents dispersed in carefully weighed trays, and then allowed to reach an equilibration weight. This procedure was necessary since the  $\text{PuO}_2$  (as received from the vendor) which was packaged under conditions of 4 percent relative humidity, was to be processed in the glove box line under an atmosphere of 30 to 60 percent relative humidity. The  $\text{PuO}_2$  was considered equilibrated when the weight became constant. The average weight gain due to moisture absorption during the equilibration period was 0.5 percent. The feed was then sampled, reweighed and again assayed as previously done before being introduced to the process line for fabrication. This program, designated as the "multiple measurement" program, involved many individual measurements. A summary of these measurements is shown in Table 2. The twelve values of plutonium content shown in Table 2 were based on various averages



and combinations of these measurements. Many of these measurements were replicate determinations on the same material or measurements on sampled feed cans. As originally proposed, the multiple measurement program was to emphasize calorimetry and coulometry measurements of feed material after this material was equilibrated. Neutron counting was added to gain additional experience in this field. Neutron counting results shown on Table 2 agree favorably with calorimetry and coulometry results; however, this may be misleading since canned feed material of known Pu content (as determined by coulometry) was used as the standard in these measurements. In addition to the above, and at the request of the AEC, feed samples were sent to the New Brunswick Laboratory for independent measurements. These results, shown as items 4, 5 and 6 on Table 2, compare favorably with results of measurements made by GE-VNC.

The value for the plutonium feed, which was used in the material balance, was obtained from the equilibrated weight and chemical assay. (Derived from the statistical average of 16 individual determinations made on 8 samples, see measurement values 2 and 3 in Table 2.)

Table II

## ISE Campaign 2 Plutonia Feed Measurements

Measurement	Measurement Method	Individual Analysis	Pu Content, %	Pu, gm
1. ARHCO <sup>(a)</sup> (1970)	Potentiometric	3	85.71	2452.0 <sup>(b)</sup>
2. GE <sup>(c)</sup> (1970)	Coulometry (Composite)	4	85.23	2448.9 <sup>(d)</sup>
3. GE (1970)	Coulometry	12	84.93	2441.2 <sup>(d)</sup>
4. NBL <sup>(f)</sup> (1970)	Potentiometric	3	85.26 <sup>(g)</sup>	2449.8 <sup>(d)</sup>
5. NBL (1970)	Potentiometric	3	85.34 <sup>(h)</sup>	2441.4 <sup>(b)</sup>
6. NBL (1970)	Potentiometric	3	85.47 <sup>(i)</sup>	2445.1 <sup>(b)</sup>
7. GE (1970)	Calorimetry (1970 ARHCO Isotopic)	8	85.97 <sup>(j)</sup>	2459.5 <sup>(b)</sup>
8. GE (1970)	Calorimetry (1970 GE Isotopic)	8	85.56 <sup>(j)</sup>	2429. <sup>(b)</sup>
9. GE (1970)	Gross Neutron (1970 ARHCO Isotopic)	20	84.65 <sup>(j)</sup>	2439.5 <sup>(b)(k)</sup>
10. GE (1970)	Gross Neutron (1970 GE Isotopic)	20	84.77 <sup>(j)</sup>	2443.0 <sup>(b)(k)</sup>
11. GE (1970)	Coincidence Neutron (1970 ARHCO Isotopic)	20	84.17 <sup>(j)</sup>	2426.9 <sup>(b)(k)</sup>
12. GE (1970)	Coincidence Neutron (1970 GE Isotopic)	20	84.29 <sup>(j)</sup>	2429.3 <sup>(b)(k)</sup>
(a) Atlantic Richfield Hanford Company		(f) New Brunswick Laboratory		
(b) Net weight dry plutonia — 2860.8 grams (ARHCO)		(g) Analyzed as—received in 52% relative humidity		
(c) General Electric		(h) Analyzed as—received in a dry box—relative humidity < 0.1%		
(d) Net weight equilibrated plutonia — 2373.3 grams (GE)		(i) Analyzed after equilibration in a dry box—relative humidity < 0.1%		
(e) Includes a —0.09 weight difference		(j) Value calculated from net and plutonium weights		
		(k) Corrected for samples utilizing measurement 3.		

## B. Product Material

Product material consisted of two sets of fuel rods, ten in each set. One set contained solid pellets, the other annular pellets with plutonium concentration slightly higher than that of the solid pellets. Samples were taken from the boats containing sintered pellets for wet chemistry plutonium content assay. Sampling plan used was specified in the GE contract, one pellet from each fourth boat.

A total of 31 assays were made, 15 for 2.58% enrichment and 16 for 2.38% enrichment. Since pellets within a given enrichment were made from the same master blend an average plutonium concentration value was determined and used in calculating the plutonium content of the sintered oxide product. The distribution of values from these measurements was used to calculate the limit of error associated with the average Pu concentration value for the specific enrichment. The total percent Limit of Error\* for each enrichment was then determined by combining the Pu concentration LE and the percent weight LE.\*\*

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\*

LE defined as boundaries within which the true value of the parameter being measured lies with a probability of 95 percent. Measurements error and sampling error are assumed to be normally distributed.

\*\*

See Reference 5.

### C. Scrap and Measured Discards

The measured discards (waste) and scrap packages were neutron coincidence counted. The procedures used were essentially the same as those followed in Campaign 1. The counting times were the same, 10 minutes, independent of whether the material was low level waste or recoverable scrap. The calibration program for this measurement consisted of counting the background and standards and fitting a curve to the corrected standards values and their associated Pu 240 equivalent values. This curve (straight line) has an intercept and a slope, each of which has an error associated with it. Some of the standards used were the same as were used for calibration during the measurement of Campaign 1 materials. In addition, three other standards utilizing 80 percent fissile plutonium were incorporated into the calibration curves.

As experienced in Campaign 1, the neutron counting equipment posed problems of instability during the measurement of Campaign 2 waste and scrap. Therefore, several calibration curves and sets of parameters had to be developed. Backgrounds differ with the time of day and the activity, and this can affect the low level waste measurements. Another uncertainty and potential problem was introduced into the system by attempting to count scrap packages which contained greater amounts of plutonium than the standards that were used to develop the calibration curve. In these



cases, the calibration curve was linearly extrapolated. No additional uncertainty was included to cover this procedure. The LE associated with the scrap and waste measurements, therefore, does not include uncertainties due to the extrapolation procedure.\*

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\*

A more detailed explanation of these measurements is given in Reference 8.



### III. Material Balance Analysis and Results

#### A. Material Measurements

##### 1. Calorimetry

Feed material that was used in the process line for fabrication of Campaign 2 fuel was measured by replacement calorimetry. Total plutonium content was determined to be 2429 grams. The limit of error associated with this measurement amounted to 191 grams plutonium or 7.8%. Although this LE is considered high, it must be noted that only a negligible portion (.05%) is due to the calorimeter. The balance is due to the high uncertainties associated with the isotopic content of the plutonium sample. The effect of isotopic analytical errors on replacement calorimetry assay results were determined.

The contributions for specific isotopes are:

<u>Isotope</u>	<u>Abundance</u> %	<u>Error</u> %	<u>Propagated Error</u> % (b)
Pu 238	0.249	10	2.66
Pu 239	75.65	0.04	0.01
Pu 240	18.48	0.1	0.03
Pu 241	4.50	0.2	0.01
Pu 242	1.13	0.4	0.00
Am 241	0.860	27.2 (a)	<u>5.06</u>
Total systematic error			7.77%

(a) ARHCO Value-used because ARHCO abundance value for Am 241 was used. All other uncertainty values

\*Reported as shown in GE Fourth Quarterly Report on ISE, GEAP-12114-7

shown are GE-VNC values.

- (b) Propagated error contribution = percent heat contribution times the error.

The feed value obtained by the calorimetric measurement method agrees closely with that obtained by the wet chemistry method (2429 vs. 2425 grams). The wet chemistry value was used to calculate the material balance because of its smaller limit of error.

2. Analytical Chemistry. As noted in the discussion of feed measurements in this report the analytical chemistry value for equilibrated feed material was used to characterize the Feed.

3. Neutron Coincidence Counting. Neutron coincidence counting was used to measure measured discards, dirty sintered, dirty green and clean green scrap. NBS limit of error values include the following systematic errors due to the following:

(1) Conversion ratios from gPu 240 equivalent units to gPu includes:

(a) Isotopic abundance uncertainties for Pu 238, Pu 240 and Pu 242 and

(b) spontaneous fission rate uncertainties for the same three isotopes.

(2) Percent uncertainty in standards -- this due to low number of standards from different blends used for calibration purposes.

B. Material Balance - Results. The material balance, which includes measurements of all streams with the exception of the absolute filters, is shown in Table 3. Absolute filter results were not used since counting procedures were not dependable.

Table 3 - Material Balance

Input

Feed by chemical assay	2425.1 ± 5.8 gPu
------------------------	------------------

Output

- |   |                  |
|---|------------------|
| <ul style="list-style-type: none"> <li>● Chem Assay Determination</li> <li style="padding-left: 20px;">Product</li> <li style="padding-left: 20px;">Clean Sintered Scrap</li> <li style="padding-left: 20px;">Sintered Samples</li> </ul>   | 2256.9 ± 7.5 gPu |
| <ul style="list-style-type: none"> <li>● Non-destructive Determination</li> <li style="padding-left: 20px;">Measured Discards</li> <li style="padding-left: 20px;">Other Scrap</li> <li style="padding-left: 40px;">Dirty Sintered</li> <li style="padding-left: 40px;">Dirty Green</li> <li style="padding-left: 40px;">Clean Green</li> </ul> | 158.8 ± 10.8 gPu |
| <ul style="list-style-type: none"> <li>● Samples - Green Mixed Oxide</li> </ul>   | 0.1 ± 0.0 gPu    |

$$\text{MUF} = \text{Input} - \text{Output} = 9.3 \text{ gPu}$$

$$\text{LEMUF} = \left\{ \text{LE}(\text{INPUT})^2 + \text{LE}(\text{OUTPUT})^2 \right\}^{\frac{1}{2}} = 14.4 \text{ gPu}$$

$$\text{MUF} = 0.38 \text{ Percent of throughput (gms. Pu Feed)}$$

All above LE values are independent.

Note - The L.E. values associated with quantities which are related cannot be combined using the simplified error propagation formula for unrelated quantities. The reason for this is that the covariance terms, which indicate the degree that quantities are related to each other, are not zero and therefore contribute to the total LE. See reference 5 for details.



#### IV. Nested Design Experiment

The analysis made on the results of the following described experiment was performed by the GE staff and the report herein was an excerpt from the initial GE draft. Values and findings are reported as offered by GE, and they apply only to the results of this experiment. It is the NBS ISE Representative's opinion that if the NDT equipment had been tailor-made for the samples used, the results and findings would be different.

A. Background and Objectives. The Integrated Safeguards Experiment has shown the importance of measuring all streams in a plutonium process line in order to assure a true material balance. The uncertainties associated with the assay methods used must also be determined and it is most important that the magnitude of these uncertainties be known so that the most appropriate methods may be applied to the problem of safeguards control.

During the latter part of Campaign 2 this subject was aired in a meeting between representatives of GE-VNC and the ISE-technical representatives which resulted in a special experiment conducted by GE-VNC to evaluate measurement errors in several Pu assay methods. Information reported herein is extracted from the results of this experiment. A detailed report on this experiment will be issued by GE.

As originally planned, a replicate sample--replicate analysis program was set up to compare the plutonium content of single pellets measured by analytical chemistry (controlled potential coulometry), passive neutron assay, and replacement calorimetry.\* Before initiation of routine measurements, tests were conducted on a single pellet to determine the precision of the planned non-destructive assay techniques. The observed percent limit of error based on ten assay determinations was 1.9 and 1.8 for gross and coincidence counts, respectively. Similar data for replacement calorimetry based on 5 determinations resulted in percent limit of error of 12.4. While this value is out of proportion to that observed for the other NDT methods, it should be pointed out that this is not indicative of calorimeter precision. The calorimeter available to GE for these tests was not designed to detect the heat generated by a single pellet sample, which was in the milliwatt range, and therefore the readings were not dependable. Because of this, the calorimetry NDA was deleted from the test and passive gamma measurements were substituted for them.

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Also, since little was known about the several variance components which make up the total Pu concentration uncertainty, another phase of this experiment was planned to identify, segregate and determine the magnitude of those components.

B. Definition of terms used in conjunction with the nested design experiment.

Random Measurement Error	The component of random error in a measuring process due to chance alone. It does not include systematic errors introduced by sampling, miscalibration or other sources.
Random Measuring Order	In this test, the measuring sequence for each sample determined by the order in which numbers were selected from a random number table. A different sequence was used for each measurement technique.
Process Pellets	Whole pellets selected and analyzed during the fuel fabrication process for product certification.
Percent Relative Error	The standard deviation of the random measurement error on Pu concentration for a single determination divided by the mean Pu fraction and expressed as a percent.
Treatment	The method of preparing pellet samples to achieve physical forms having varying degrees of homogeneity. From most homogeneous to least, these include (1) pulverized, (2)

halved, and (3) whole pellets.

#### Experiment Pellets

Pellets selected at the same time and from the same firing boats as process pellets but analyzed at a later date after the special sample treatment required in the Nested Design.

Of the two passive counting methods investigated, gamma counting with NaI (Tl) detectors had smaller error than neutron counting (0.8% versus 1 to 2% relative), but the gamma counting method was very sensitive to the sample configuration.

Within-pellet variation and between-boat variation were found to be insignificant. Between-pellet variation was as large as the dissolution variance. The analytical variation was approximately half that of the dissolution variance.

C. Description of Method. The basic experimental plan is known as a balanced, staggered nested design. It is a balanced design because the test is symmetrical with respect to a repeating pattern of pellet treatments; it is a staggered design because some lower level replicate measurements were not made. This was done to reduce the cost of the experiment without sacrificing any of the estimates of the variance components. It is a nested design because the lower order terms in the statistical model are combined with successively higher levels until they account for the total measurement error. In this test, the five levels (from low to high) were:



- Variability between aliquants of the same sample solution (called the "aliquanting error" and include coulometer errors)
- Variability between solutions made from pulverized samples (called the dissolution error)
- Variability between the analysis of halves of the same pellet (called pellet inhomogeneity)
- Variability between individual pellets from the same firing boats (called between-pellet error)
- Variability between means of firing boats (called between-boat errors)

The test design involved two classes of pellets. The process class of pellets included the samples normally analyzed for  $\text{Pu}/(\text{Pu}+\text{U})$  to certify the product. Process sample pellets were withdrawn from the same boats as the experimental pellets but were analyzed in the Spring of 1971 at the time the fuel was fabricated. Although the experimental class of pellets was withdrawn at the same time as the process pellets, they were subjected to special treatments and additional testing before being measured in the fall of 1971.

Three pellet treatments were defined as follows:

1. Whole pellets - significant differences between pellets would suggest differences within the batch.

2. Halved pellets - significant differences between pellet halves from the same pellet would indicate inhomogeneity within a pellet.
3. Pulverized pellets - pulverization should assure homogeneous samples.

#### D. Findings

General. In comparing controlled potential coulometry to various counting techniques, it is important to note that the relative error for the chemical method is for the weight ratio of plutonium to oxide and the relative error for a non-destructive counting method (NDA) is for the weight ratio of certain plutonium isotopes to oxide. Thus, the relative errors are not directly comparable, since the chemical relative errors do not include the uncertainty in the factor for converting the weight of the isotopes measured by NDT to plutonium weight.

The use of suitable isotopic standards would allow NDT values to be judged for accuracy. In the absence of suitable standards, the methods studied can be ranked with respect to precision for determining  $\text{Pu}/(\text{UO}_2 + \text{PuO}_2)$  by dividing the methods into three groups of about equal precision.

Most precise:	Controlled potential coulometry
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Next most Precise:	Non-destructive gamma counting at 0.414 MeV for whole pellets
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Non-destructive gamma counting  
at 0.208 MeV for whole pellets.

Less precise:

Non-destructive gross neutron  
counting with any pellet treatment.

Least precise:

Non-destructive gamma counting at  
0.414 MeV for both halved and  
pulverized pellets.

Non-destructive gamma counting for  
the sum of the 0.208 and 0.416 MeV  
using halved and pulverized pellets.

Non-destructive coincident neutron  
counting for any pellet treatment.

Non-destructive gamma counting at  
0.208 MeV for halved and pulver-  
ized pellets.

As pointed out in Section IV, the calorimetric re-  
sults were not indicative of the accuracy of the instrument  
since it was not designed to measure heat from a single pellet,  
the same rationale applies to the neutron and gamma counting  
measurements used here since the experimental configuration and  
procedure (time, counting statistics, etc.) were not compatible  
and direct comparison was not realistic.

## Non-Destructive Counting Techniques

1. The relative errors for the best counting technique, in this experiment (the gamma 0.414 MeV measurement), are larger than for the controlled potential coulometric determination of Pu/(UO<sub>2</sub>+PuO<sub>2</sub>) (0.7 versus 0.28).
2. There was a significant difference in the percent relative errors between the measurement techniques for the various pellet treatments designed to measure fuel homogeneity within a sample. As expected, the neutron measurements were affected least by the pellet treatments and the gamma measurements were affected the most.
3. For gamma counting methods, the within-pellet relative error is smaller for whole pellets than for either halved or pulverized pellets.
4. For gamma counting, halved and pulverized pellets had greater variability between pellets than within pellets. Thus, variability in preparation was a major source of the measurement error. Since whole pellets showed consistently less variability between pellets than within pellets, it was concluded pellet preparation is not a major source of error for whole pellets.
5. For gross and coincident neutron counting, the relative errors for both within and between pellets do not depend on the three types of pellet treatments.



6. The gross neutron counting method has appreciably smaller relative errors on homogeneous mixed oxide fuel (within, between and total) than does the neutron coincident counting method.

#### Analytical Chemistry

1. The Pu/(UO<sub>2</sub>+PuO<sub>2</sub>) analysis by controlled potential coulometry had the smallest relative percent error of any method of determination (0.28).
2. The relative error in the Pu/Pu+U determination was 2 times the relative error for the Pu/(UO<sub>2</sub>+PuO<sub>2</sub>) determinations (0.54 versus 0.28%).
3. The measurement errors are essentially divided between "aliquanting" and "pellet dissolution." This means that significant improvements in both aliquanting and sample preparation must be made in order to improve the overall analytical measurement capability.
4. Pellet inhomogeneity, as measured by differences between pellet halves and between pellets in the same boat, was too small to be measured, in comparison with other sources of variation within the experiment. This suggests that the fuel is homogeneous with respect to plutonium enrichment as measured by the tested controlled coulometric method.
5. A relative measurement error of 0.06% was observed for 9

National Bureau of Standards samples (calibration standards).

6. A bias of 0.015% relative in the  $\text{Pu}/(\text{Pu}+\text{U})$  determinations was observed between process and experimental samples. It is believed that this difference was due to uncontrolled changes within the analytical process between the spring and fall of 1971.

#### E. Summary - Nested Design

The test consisted of 15 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 non-destructive gamma and neutron counting methods. These same pellets were then chemically dissolved and measured by the controlled potential coulometric method.

Total % CV for the experimental pellets was calculated to be 0.28%, as determined by the controlled potential coulometric method for  $\text{Pu}/(\text{UO}_2+\text{PuO}_2)$ , and a bias of 0.015% relative was observed between the process and experimental sample results. This bias reflects differences between two analysis periods.

The variance components which contributed to the total % CV are as follows:

(1)	aliquanting error . . . . .	0.11%
(2)	dissolution error . . . . .	0.18
(3)	pellet inhomogeneity error. . . . .	0.00
(4)	between pellet error. . . . .	0.18
(5)	between boat error. . . . .	0.07

## V. Campaign 2 - Summary of Findings

It was stated in the introduction of this report that the Campaign 2 experimental plan followed closely that of Campaign 1, differing only in several areas where Campaign 1 experience indicated that refinements or changes should be made. These changes were mainly in the calibration of equipment and material handling and treatment areas. The result of the Campaign 2 material balance ( $LEMUF=0.6\%$  of throughput) shows a significant improvement over that of Campaign 1 ( $4.0\%$  of throughput). This accomplishment is primarily attributed to the inclusion of the equilibration treatment of the  $PuO_2$  feed, which resulted in more accurate chemical assay data since samples from the equilibrated powder were truly representative of the bulk material.

One of the features added to Campaign 2 was provision of longer blending time in the feed preparation. It was expected that this treatment would ensure maximum homogeneity of the blended oxides, and hence a lower percent coefficient of variation associated with the Pu content. The results of such treatment were not, however, as expected. Blending time for two master blends was essentially the same, but the % CV

varied considerably, as indicated below:

<u>Master blend</u>	<u>%CV</u>	<u>n</u>
MO100	0.43	15
MO101	1.13	16

There was no apparent explanation for this behavior and during the conduct of Campaign 2 it appeared logical to attribute the difference to pellet inhomogeneity or to lack of complete dissolution before chemical analysis. This matter was somewhat enlightened by the results of the nested design experiment conducted after completion of the Campaign. The experimental pellets used in the nested design experiment produced a total % CV of .28. This is in contrast to the total % CV calculated for the process pellets (from the same lot as the experimental pellets) of .43. A possible explanation, after observing this substantial difference is that the smaller analytical and dissolution variances were the result of more careful and precise handling in the case of the experimental pellets.

The nested design experiment was successful in that it revealed an acceptable estimate of the variability inherent in the controlled potential coulometric method of measurement of mixed Pu and U oxides. This variance compares favorably with results of other laboratories when conducting similar tests. (6,7) It also characterized the various NDA methods of measuring Pu content of materials in various forms within the fabrication cycle and therefore will assist nuclear



fuel manufacturers in selecting and utilizing NDA equipment where it can best be applied.

Effective utilization of calorimetry can be made if accurate data is known on the isotopic content of the material measured. Calorimeter results compare very favorably with chemical analysis results. It is important to note, however, that the calorimeter must be designed to operate effectively over the anticipated heat output range.

Campaign 2 has shown that extra care in handling, recording, blending and measuring material in the process line will improve material control within the plant. This can be done with little additional cost, and the net result may be a monetary gain to the manufacturer.

This campaign has shown that much may be accomplished by non-destructive counting techniques; however, their accuracy depends heavily upon carefully prepared standards. In this campaign, standards used in neutron counting techniques contained plutonium from sources other than that of the product being measured. This created variances which were reflected in the results and, if not accounted for in a material balance, could be a source of error.

## VI. Conclusions

At the completion of Campaign 1 there was evidenced a strong need for tailoring operational procedures which would standardize methods of sampling, measurement, calibration, reporting, material balance and limit of error calculations. This was so reported in NBS Report No. 10 660. This need is even more strongly evidenced by the results of Campaign 2. Experience gained during the Integrated Safeguards Experiment indicates that such action could be taken without imposing much hardship upon the manufacturer. Such action is necessary to assure a common base for material control in all facilities in the nuclear fuel field. It would also eliminate many variables confronting the government inspector during a periodical inspection.

As was experienced in Campaign 1, the application of non-destructive testing techniques made the closed measured material balance in Campaign 2 possible. The results of the nested design experiment indicates that the counting techniques tested have relative measuring errors in the range of 1 to 2 percent. These determinations were made by highly trained engineers at GE, and under carefully prepared conditions. In a production environment, however, where working standards may not be carefully prepared and/or where operating personnel may not be conscious of the intricacies of

these techniques, the range of errors could be much greater. Even though this type of equipment will be refined, with improved accuracy and precision as the equipment is tailored for specific tasks, it is unlikely that NDT methods, as used in the ISE, will replace analytical chemical determinations for certain applications. It appears certain that the application of NDA will have increasing use for process control and the measurement of heterogeneous materials. When considering the utilization of NDA in the nuclear fuel fabrication cycle, a judicious choice of equipment, consistent with requirements of accuracy, contribution to the LE of MUF, cost of test, and type of material to be measured, must be exercised.

## VII. Acknowledgments

The ISE experiment was conducted in a commercial facility in a production environment. Unlike Campaign 1, where there was much planning and discussions and interference with production by the ISE technical representatives, Campaign 2, under the pressure of delivery of product to the customer, was performed almost exclusively by General Electric Company people. In particular, GE was responsible for the conduct of the "Nested Design Experiment."

As was the case with the Campaign 1 report, the data used for the preparation of the Campaign 2 report was a collaborative effort by NBS, TSO (Brookhaven) and GE-VNC. Much of the data shown in the report was taken from the GE Fourth Quarterly Report of the Integrated Safeguards Experiment, GEAP-12114-7. Also, the brief account on the nested design experiment contained in this report was extracted from the GE-VNC draft report entitled, "Measurement Errors in Several Pu Assay methods Based on a Balanced Staggered Nested Design." A final, detailed report on this work will be issued by GE as GEAP-12283.

We wish to acknowledge the assistance provided by the TSO (Brookhaven) Staff. This report reflects a number of opinions and thoughts of Sylvester C. Suda and William Marcuse, authors of the paper, "Integrated Safeguards Experiment at a Plutonium Fuels Laboratory." (See reference 3.)



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