DETAILED ANALYSES OF MEASUREMENTS
IN THE
INTEGRATED SAFEGUARDS EXPERIMENT

This paper is based on work performed under
United States Atomic Energy Commission
Contract No. AT(49-2)-1165
NATIONAL BUREAU OF STANDARDS

The National Bureau of Standards¹ was established by an act of Congress March 3, 1901. The Bureau's overall goal is to strengthen and advance the Nation's science and technology and facilitate their effective application for public benefit. To this end, the Bureau conducts research and provides: (1) a basis for the Nation's physical measurement system, (2) scientific and technological services for industry and government, (3) a technical basis for equity in trade, and (4) technical services to promote public safety. The Bureau consists of the Institute for Basic Standards, the Institute for Materials Research, the Institute for Applied Technology, the Center for Computer Sciences and Technology, and the Office for Information Programs.

THE INSTITUTE FOR BASIC STANDARDS provides the central basis within the United States of a complete and consistent system of physical measurement; coordinates that system with measurement systems of other nations; and furnishes essential services leading to accurate and uniform physical measurements throughout the Nation's scientific community, industry, and commerce. The Institute consists of a Center for Radiation Research, an Office of Measurement Services and the following divisions:

THE INSTITUTE FOR MATERIALS RESEARCH conducts materials research leading to improved methods of measurement, standards, and data on the properties of well-characterized materials needed by industry, commerce, educational institutions, and Government; provides advisory and research services to other Government agencies; and develops, produces, and distributes standard reference materials. The Institute consists of the Office of Standard Reference Materials and the following divisions:

THE INSTITUTE FOR APPLIED TECHNOLOGY provides technical services to promote the use of available technology and to facilitate technological innovation in industry and Government; cooperates with public and private organizations leading to the development of technological standards (including mandatory safety standards), codes and methods of test; and provides technical advice and services to Government agencies upon request. The Institute also monitors NBS engineering standards activities and provides liaison between NBS and national and international engineering standards bodies. The Institute consists of the following divisions and offices:

THE CENTER FOR COMPUTER SCIENCES AND TECHNOLOGY conducts research and provides technical services designed to aid Government agencies in improving cost effectiveness in the conduct of their programs through the selection, acquisition, and effective utilization of automatic data processing equipment; and serves as the principal focus within the executive branch for the development of Federal standards for automatic data processing equipment, techniques, and computer languages. The Center consists of the following offices and divisions:

THE OFFICE FOR INFORMATION PROGRAMS promotes optimum dissemination and accessibility of scientific information generated within NBS and other agencies of the Federal Government; promotes the development of the National Standard Reference Data System and a system of information analysis centers dealing with the broader aspects of the National Measurement System; provides appropriate services to ensure that the NBS staff has optimum accessibility to the scientific information of the world, and directs the public information activities of the Bureau. The Office consists of the following organizational units:

¹ Headquarters and Laboratories at Gaithersburg, Maryland, unless otherwise noted; mailing address Washington, D.C. 20234.
² Part of the Center for Radiation Research.
³ Located at Boulder, Colorado 80302.
DETAILED ANALYSES OF MEASUREMENTS
IN THE
INTEGRATED SAFEGUARDS EXPERIMENT

C. N. Smith
F. J. Perella
Technical Analysis Division
National Bureau of Standards

This paper is based on work performed under
United States Atomic Energy Commission
Contract No. AT(49–2)–1165

IMPORTANT NOTICE

Approved for public release by the
Director of the National Institute of Standards and Technology (NIST)
on October 9, 2015.
REPORT ON THE
DETAILED ANALYSES OF MEASUREMENTS
IN THE
INTEGRATED SAFEGUARDS EXPERIMENT

Prepared by
C. N. Smith
Physicist

F. J. Perella
General Engineer

Reviewed by:
Walter G. Leight
Richard T. Penn, Jr.
George Suzuki

Approved by: W. Edward Cushen, Chief
Technical Analysis Division
## CONTENTS

<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>Preface</td>
<td>1</td>
</tr>
<tr>
<td>Introduction</td>
<td>3</td>
</tr>
<tr>
<td><strong>Feed Material</strong></td>
<td></td>
</tr>
<tr>
<td>Campaign 1 - Make-up and Treatment</td>
<td>6</td>
</tr>
<tr>
<td>Campaign 2 - Make-up and Treatment</td>
<td>8</td>
</tr>
<tr>
<td>Feed Results</td>
<td>10</td>
</tr>
<tr>
<td>Summary on Feed</td>
<td>11</td>
</tr>
<tr>
<td><strong>Product Material</strong></td>
<td></td>
</tr>
<tr>
<td>Discussion on Campaign 1</td>
<td>12</td>
</tr>
<tr>
<td>Discussion on Campaign 2</td>
<td>15</td>
</tr>
<tr>
<td>Product Results</td>
<td>17</td>
</tr>
<tr>
<td>Summary on Product</td>
<td>17</td>
</tr>
<tr>
<td>Additional Comments</td>
<td>17</td>
</tr>
<tr>
<td><strong>Scrap and Measured Discards</strong></td>
<td></td>
</tr>
<tr>
<td>Discussion on Campaign 1</td>
<td>20</td>
</tr>
<tr>
<td>Discussion on Campaign 2</td>
<td>25</td>
</tr>
<tr>
<td>Summary on Scrap and Waste</td>
<td>26</td>
</tr>
<tr>
<td><strong>Summary</strong></td>
<td></td>
</tr>
<tr>
<td>Feed Material - PuO₂</td>
<td>27</td>
</tr>
<tr>
<td>Product Material - Sintered Mixed Oxide</td>
<td>27</td>
</tr>
<tr>
<td>Scrap and Measured Discards</td>
<td>27</td>
</tr>
<tr>
<td>Overall Balance</td>
<td>28</td>
</tr>
<tr>
<td>References</td>
<td>29</td>
</tr>
</tbody>
</table>
PREFACE

An Integrated Safeguards Experiment (ISE) conducted within a nuclear fuels manufacturing plant is, in a broad sense, the tracing of fissile material through every stream in a process or processes under production conditions in an attempt to uncover those factors which contribute to the imbalance (specifically the material unaccounted for (MUF)) when a material balance for such plant is periodically calculated. The ISE, discussed in this paper, had as its primary objective the evaluation of the usefulness to Safeguards of material balance accounting based on all measurements being completely quantitative. This was accomplished by the utilization of non-destructive testing techniques (NDT) to measure difficult to measure materials. Generally, the fissile material content of these hard-to-measure materials was previously determined by either an educated guess or by a "by-difference" conclusion.

The ISE, from which material for this paper was drawn, will be reported in detail in a report to be issued by the USAEC. In conducting the experiment, however, many problems were encountered which required detailed effort not anticipated in the planning of the project. These details are considered important enough to be reported separately in an effort to emphasize their significance. The intent of this paper, therefore, is to acquaint those interested
in possibly conducting similar integrated tests of some of the details and problems associated with experimental determination of plutonium content of the major flows of a mixed-oxide fabrication process.
INTRODUCTION

In January 1968, the U.S. Atomic Energy Commission (AEC), Office of Safeguards and Materials Management established a Safeguards System Studies program which has as a primary objective the study and analysis of the various nuclear fuel manufacturing processes, for the purpose of developing a basis upon which criteria for acceptable limits on material unaccounted for (MUF), normal operating losses (measured discards), shipper-receiver (S/R) differences and unmeasured special nuclear material (SNM) inventory could be established. Necessary information was to be compiled from both historical and current plant files and from it develop analytical models which could be used to evaluate plant data leading to the establishment of criteria for acceptable limits for MUF, measured discards, S/R differences and other critical factors. Visits to licensee plants soon established the fact that historical data was lacking in quality and depth and therefore not applicable to the establishment of criteria. Also, current data was not applicable since much of it was based on questionable measurement data.

As a result of the findings of this early industry survey the AEC decided to conduct Integrated Safeguards Experiments (ISE) within licensee plants which were producing nuclear fuel materials of major critical interest from a need-to-safeguard viewpoint.
The Integrated Safeguards Experiment was conducted at the General Electric Company Plutonium Laboratory, Vallecitos Nuclear Center near Pleasanton, California. This laboratory contains a fabrication process where plutonium dioxide and uranium dioxide are physically mixed to produce mixed oxide master blends of a given enrichment (percent of plutonium in the mixed oxide); the material is then prepressed, granulated, pressed, sintered, and acceptable pellets loaded into fuel clad. The ISE consisted of two campaigns of mixed oxide material: one utilized 90 percent fissile plutonium and the other 80 percent. Each campaign consisted of two different enrichments, with each enrichment made up of one or more master blends. The higher of the two enrichments within a given campaign was fabricated into annular pellets, while the material from the lower enrichment was formed into solid pellets. The measurements associated with the major flows of the fabrication process are analyzed for the two jobs (campaigns) which were run as part of the experiment. The major flows considered for this paper are the plutonium dioxide feed, the sintered mixed oxide product, and the scrap and measured discards (waste) materials. For each of these flows the details of each campaign are discussed; results are presented for the various plutonium analysis techniques which were applied. In each section, the procedures developed for determining the uncertainties and for combining the random
and systematic errors to obtain a total limit of error are discussed or referenced.

In addition, this paper contains a few comments and observations on the subject of plutonium concentration uncertainties in the propagation of errors; specifically, it deals with the coefficient of variation (CV) and limit of error (LE) values for the various blends of product.
FEED MATERIAL

Feed Material for each campaign is discussed separately in order to identify the difference in handling procedures.

Campaign 1 - Make-up and Treatment

The makeup of the feed material used in Campaign 1 was rather complex. The plutonium dioxide used in this first campaign was obtained in 1969. Six cans of PuO₂ were used; namely, cans designated as A, B, C, E, F, and G. Cans E, F and G were unopened, full cans. On the other hand, some of the contents of cans A, B and C had been used for other work in 1969. It was assumed that the atmosphere in the 3 unopened cans remained the same as when sealed by the shipper. According to information from the shipper, (ARCHO), the relative humidity (RH) of the loading box atmosphere was about 4 percent. Another assumption was that the 3 open cans (A, B and C) had picked up moisture during their use in 1969, since the RH of the GE plutonium fabrication laboratory is normally in the 30-60 percent range. Equilibration test data on plutonium dioxide powder, indicates a sharp rise in the powder weight within the first few hours when "very dry" powder is exposed to a 50% RH atmosphere. It was therefore assumed that the moisture content of the unopened cans was different than that for the opened cans of Campaign 1 feed material. This was confirmed by equilibration tests performed later. These equilibration experiments indicated that plutonium concentration in PuO₂ can change by

-6-
as much as 1 percent depending upon temperature, humidity, particle size and surface area. For Campaign 1, samples were taken from cans B, C, E and G, for wet chemistry analysis of the plutonium concentration. It was generally agreed that any PuO₂ samples sent to the plutonium analytical laboratory would reach the equilibrium plateau before they were analyzed. Therefore, any "as received" plutonium concentration values were assumed to be stable insofar as the analyzing environment was concerned. Before the samples were destructively analyzed for plutonium content, the samples were "dried" at a selected temperature to drive off moisture. According to GE, plutonium concentration values obtained under "dried" conditions were supposed to represent the dry received plutonium dioxide. It was interesting to note that the average value of the 3 sub-samples taken from a composite sample was 87.27% Pu for a "dried" condition when the drying temperature was in the 200-300°C range. The average value for these same 3 sub-samples was 88.09% for a "dried" condition when the drying temperature was in the 700-800°C range. It is clear that the meaning of the word "dried" depends on conditions, and that it must be defined by a specific temperature range which will condition the sample in such a way that it may be considered representative of the feed. Since the drying process used by the plant to produce feed that was representative of the total lot could not be supported quantitatively, the results from
chemical assay were not considered usable in measured material balance calculations.

The nondestructive technique of replacement calorimetry was used for the measurement of plutonium content of the PuO₂ feed material. All cans of material used in Campaign 1 were measured at least once. The limit of error (LE) associated with the feed gPu value was due mainly to the uncertainty associated with the contained Pu 238 and Am 241. The GE Plutonium Laboratory did not have adequate analytical capability for measurement of Am 241; therefore, the vendor's value (corrected for growth) and uncertainty had to be used in the calculation of the grams plutonium as determined by calorimetry. The LE associated with the gPu in the feed material was 4% for Campaign 1. (For details on the procedure for calculating the total LE associated with calorimetrically determined values, see reference 1).

Campaign 2 - Make-up and Treatment

The PuO₂ feed material was first calorimetrically determined which revealed an error in the vendor's isotopic values, then equilibrated, sampled and analyzed coulometrically, reweighed, calorimetrically determined again and finally introduced to the clean process line for fabrication purposes. The 80% fissile plutonium dioxide was contained in three cans. The contents of each can and the accompanying composite sample were equilibrated for several days. This equilibration process consisted of allowing the material to pick up moisture
from the glove box system until there was no additional weight change. This was accomplished by transferring the contents of the PuO$_2$ cans to flat pans permitting a greater surface of the oxide to be exposed to the glove box atmosphere. The change in weight averaged about 0.5 percent; this value representing a minimum since there was a lapse of time between the opening of the shipping containers and the transferring to the large tared equilibrating pans.

Relative humidity (RH) gauges indicated that the atmosphere in the analytical laboratory was in approximately the same range as that of the fabrication laboratory. Thus, this equilibration process eliminated or, at least, minimized a variable which Campaign 1 feed material was subjected to suggesting questionable results, and allowed for a meaningful value and LE to be obtained for Campaign 2 feed material by an accurate chemical analysis method.

After equilibration, eight samples were withdrawn for Pu analysis using the coulometry technique: 2 from each of the three cans as well as from the composite. The average of the 8 plutonium concentration values was used for the feed concentration value for the purposes of the material balance. The distribution of values was also used to calculate the limit of error associated with the mean concentration value. No additional uncertainty (e.g., due to moisture difference between fabrication and analytical laboratories) except for that due to weighing, was included.
in the computation of the feed material LE value using chemical assay information. (See reference 1 for LE formulation details.)

The three cans of equilibrated PuO$_2$ material were removed from the fabrication line and each measured twice by calorimetry. The uncertainty of the calorimetry measurements was due to the uncertainty associated with the Pu 238 and Am 241 abundance determinations. As with Campaign 1, the vendor's value for Am 241, corrected for buildup, had to be used since the plant did not have that capability on a routine basis. The LE associated with the gPu value in the feed material was

$\sim$8\% for Campaign 2. This is twice as great as that in Campaign 1 and it reflects the percentage increase in the heat contribution from Pu 238 and Am 241 for 80\% fissile plutonium.

**Feed Results**

<table>
<thead>
<tr>
<th>Technique</th>
<th>Campaign 1</th>
<th>Campaign 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chemical Assay</td>
<td>NA</td>
<td>2425.1 ± 5.8 gPu</td>
</tr>
<tr>
<td>Calorimetry</td>
<td>4919. ± 197. gPu</td>
<td>2429. ± 190. gPu</td>
</tr>
</tbody>
</table>
Summary on Feed

1. The moisture problem which obscured true Campaign 1 feed values was solved in Campaign 2 by equilibrating the PuO₂ feed. Therefore an improvement over Campaign 1 was made and good chemical assay data was obtained for Campaign 2 feed.

2. No improvement was made on calorimetry. In fact, the percent LE was a factor of two larger for Campaign 2 than for 1 because the percent heat contribution from Pu 238 and Am 241 isotopic abundance was higher for the 80% fissile material.

3. There was remarkably good agreement between the chemical assay and calorimetry results for Campaign 2 feed.
Discussion on Campaign 1

There were two master blends of material made up for each of the two enrichments. The two master blends for a given enrichment were supposed to be the same. For the first enrichment, master blends M0094 and M0095 appeared to be statistically different according to the F test* on the product plutonium concentration variances. Because of this, the grams of M0094 mixed oxide and the M0094 plutonium concentration data were kept and treated separate from the M0095 information. Details are presented below. The master blends of the second enrichment, M0096 and M0098, were not statistically different, and therefore probably came from the same material sources. Thus, for the second enrichment, all the information was pooled together.

For the M0094 material, the total grams plutonium was calculated by multiplying the average M0094 plutonium concentration value by the total grams of M0094 mixed oxide that were loaded into the rods. The percentage LE associated with the M0094 grams plutonium value was obtained by adding together the LE values (in percentages) for the average plutonium

* The F statistic is the ratio of the variances of the two samples. The F test provides the probability that the two samples came from normal distributions with the same variance.
concentration and for the total weight of mixed oxide* The same procedure was followed for M0095. To obtain the LE on the total grams plutonium in the first enrichment, the LE values for M0094 and M0095, in absolute units, were propagated in the usual manner. The usual error propagation technique was again used to combine the two enrichment LE values to obtain the Campaign 1 total product LE. The usual propagation of error technique is applied in all situations except where the total grams of mixed oxide value is multiplied by the average Pu concentration value. The treatment of errors in this case requires a systematic procedure because a collection of values is being multiplied by a constant.

* See references 1 and 2 for discussion of the philosophy of this procedure.
The following tables give information about the Campaign 1 Product as determined by chemical assay.

Product Average Pu Concentration and LE

<table>
<thead>
<tr>
<th>Enrichment</th>
<th>n*</th>
<th>Average Pu concentration</th>
<th>LE</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 M0094</td>
<td>13</td>
<td>0.03548</td>
<td>.00020</td>
</tr>
<tr>
<td>M0095</td>
<td>12</td>
<td>0.03536</td>
<td>.00035</td>
</tr>
<tr>
<td>2 M0096, 98</td>
<td>26</td>
<td>0.03427</td>
<td>.00011</td>
</tr>
</tbody>
</table>

Product Grams of Mixed Oxide

<table>
<thead>
<tr>
<th>Enrichment</th>
<th>Grams of Mixed Oxide</th>
<th>LE</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 M0094</td>
<td>26323.2</td>
<td>0.8</td>
</tr>
<tr>
<td>M0095</td>
<td>28993.3</td>
<td>0.7</td>
</tr>
<tr>
<td>2 M0096, 98</td>
<td>61249.0</td>
<td>0.9</td>
</tr>
</tbody>
</table>

n* - Number of Production Samples.
Product gPu and LE

<table>
<thead>
<tr>
<th>Enrichment</th>
<th>Grams Plutonium</th>
<th>LE</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1959.1</td>
<td>11.5</td>
</tr>
<tr>
<td>MO094</td>
<td>933.9</td>
<td>5.3</td>
</tr>
<tr>
<td>MO095</td>
<td>1025.2</td>
<td>10.2</td>
</tr>
<tr>
<td>2</td>
<td>2099.0</td>
<td>6.8</td>
</tr>
<tr>
<td>1+2</td>
<td>4058.1</td>
<td>13.4</td>
</tr>
</tbody>
</table>

The fuel rods were gamma scanned twice; once using a NaI detector and later using a GeLi detector. Unfortunately, the analyses of the gamma scanning data were not completed in time to be used in this report. The same explanation applies to the 20 rods of Campaign 2.

Discussion on Campaign 2

The product consisted of two sets of fuel rods. One set of 10 rods contained solid pellets; the other set of 10 rods contained annular pellets whose plutonium concentration was slightly higher than that for the solid pellets.

Samples were withdrawn for purposes of plutonium concentration determination from the boats of sintered mixed oxide pellets in accordance with the fuel fabrication contract, one sample every fourth boat. Approximately 15 plutonium concentration determinations were made for each of the two enrichments. Since pellets within a given enrichment were supposed to come from the same population (master blend) an average plutonium concentration value was determined and used in determining the plutonium content of the sintered mixed oxide product. The distribution of values was also used to calculate the limit of error associated with the
average Pu concentration value for the enrichment. This LE value, converted to a percentage, was added to the percentage weighing LE to obtain the percentage LE on the product on an enrichment basis. To obtain the product LE for the campaign, the absolute LE values for each enrichment were combined using the usual error propagation techniques.

The following tables give information about the Campaign 2 Product as determined by chemical assay.

**Product Average Pu Concentration and LE**

<table>
<thead>
<tr>
<th>Enrichment</th>
<th>n*</th>
<th>Average Pu Concentration</th>
<th>LE</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 MO100</td>
<td>15</td>
<td>0.02584</td>
<td>.000062</td>
</tr>
<tr>
<td>2 MO101</td>
<td>16</td>
<td>0.02388</td>
<td>.000143</td>
</tr>
</tbody>
</table>

**Product Grams of Mixed Oxide**

<table>
<thead>
<tr>
<th>Enrichment</th>
<th>Grams of Mixed Oxide</th>
<th>LE</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 MO100</td>
<td>39260.5</td>
<td>0.8</td>
</tr>
<tr>
<td>2 MO101</td>
<td>43424.8</td>
<td>0.8</td>
</tr>
</tbody>
</table>

**Product gPu and LE**

<table>
<thead>
<tr>
<th>Enrichment</th>
<th>Grams Plutonium</th>
<th>LE</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 MO100</td>
<td>1014.5</td>
<td>2.5</td>
</tr>
<tr>
<td>2 MO101</td>
<td>1037.0</td>
<td>6.2</td>
</tr>
<tr>
<td>Campaign 2</td>
<td>2051.5</td>
<td>6.7</td>
</tr>
</tbody>
</table>

*n - Number of Production Samples.*
Product Results

<table>
<thead>
<tr>
<th>Techniques</th>
<th>Campaign 1</th>
<th>Campaign 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chemical Assay</td>
<td>4058.1 ± 13.4 gPu</td>
<td>2051.5 ± 6.7 gPu</td>
</tr>
<tr>
<td>NaI</td>
<td>Results not Available</td>
<td></td>
</tr>
<tr>
<td>GeLi</td>
<td>Results not Available</td>
<td></td>
</tr>
</tbody>
</table>

Summary on Product

1. The % LE for each campaign was 0.33%.
2. More blending work was performed on Campaign 2 master blends than on Campaign 1 materials. One conclusion may be that the plutonium heterogeneity variance between pellets does not appear to be one of the dominant variances.
3. There was a factor of ~1.7 difference between the LE associated with gPu of enrichment 1 and 2 of Campaign 1. For Campaign 2 the difference between the two enrichments was a factor of ~2.5. In one campaign the annular pellets had the larger LE, in the other campaign it was the solid pellets.

Additional Comments

As indicated in the introduction of this paper, a few observations and comments would be made on the uncertainty of the product plutonium concentration values. The distribution of n production samples (sintered pellets) produced the following coefficients of variation for the various blends:
<table>
<thead>
<tr>
<th>Campaign</th>
<th>Master Blend</th>
<th>%CV</th>
<th>n</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>MO094</td>
<td>0.93</td>
<td>13</td>
</tr>
<tr>
<td></td>
<td>MO095</td>
<td>1.57</td>
<td>12</td>
</tr>
<tr>
<td></td>
<td>MO096</td>
<td>0.90</td>
<td>14</td>
</tr>
<tr>
<td></td>
<td>MO098</td>
<td>0.61</td>
<td>12</td>
</tr>
<tr>
<td>2</td>
<td>MO100</td>
<td>0.43</td>
<td>15</td>
</tr>
<tr>
<td></td>
<td>MO101</td>
<td>1.13</td>
<td>16</td>
</tr>
</tbody>
</table>

The above data may emphasize the importance of the complete blending process. For example, we observed that a longer blending time was performed on the fourth blend of Campaign 1 (MO098) than on the first three blends. The %CV for (MO098) blend is lower than any of the others, which may be attributable to the longer blending time.

In Campaign 2, however, the preparation and blending time for both blends involved (MO100 and MO101), were approximately the same and yet the difference between the %CV of these blends is a factor of approximately 2.6. Considering the similarity of preparation and blending time of both blends, why should there be such a large difference in the %CV? Possibly the non-homogeneity of the pellet sample or the lack of complete dissolution before analysis may be a logical conclusion. In the case of blend MO100 the GE-VNC analytical laboratory rejected the results of five of the fifteen samples as unusable and requested substitute samples from this blend. These were analyzed and results recorded. This was not repeated for blend MO101 since the analyses were considered normal.
Although considerably more effort was put into the blending preparation for Campaign 2 material than there was for Campaign 1, pooled CV values and the total % LE values indicated there was little difference between the results of the two campaigns.

A nested design experiment was performed by GE-VNC using samples from blend MO100 which had a 0.43 %CV value. The objective of this experiment was to evaluate the errors between and within plutonium determinations when using three different measurement techniques. This experiment also was expected to elucidate and segregate the several variance components, such as the analytical, dissolution, Pu heterogeneity, lot-to-lot and others. The results of this experiment have not yet been reported; however, in light of the non-consistent behavior of the process, it is questionable whether results of such tests will be useful. Such results may be meaningful only if many nested design type experiments were conducted within the plant in order for the process to be characterized under its various conditions. Practical and economic considerations of this approach, however, would probably preclude doing this. Even if many sets of results were obtained, the usefulness of such information would be questionable in the propagation of error to obtain a meaningful LEMUF value. How would an inspector or plant operator know which set of results to use in the propagation? In plutonium fuel manufacturing plants where
the %CV values vary greatly between blends, the best type of information to use for LEMUF calculations is the distribution of current values. This will probably require more samples than are normally taken, but it provides a means for obtaining a meaningful LEMUF value which, after all, is the judge of the MUF value.
SCRAP AND MEASURED DISCARDS

Discussion on Campaign 1

The measured discards (waste) and scrap materials resulting from the fabrication of mixed oxide pellets for Campaign 1 were measured for plutonium content by passive neutron coincidence counting. This method detects neutrons from the spontaneous fission of isotopes such as Pu238, Pu 240 and Pu 242. The standards used for calibration purposes were fabricated from existing mixed oxide materials. Element and isotopic content from previous analyses were used. The standards were originally produced to meet the needs of the Plant Instrumentation Program (PIP).* The gPu limit of error (LE) values for several of the standards are larger than one would normally use in materials balance accounting. Another point to mention is that some of the scrap packages were "hotter" than the largest standard used in the calibration. In these cases the calibration curve was linearly extrapolated. No additional uncertainty was included to cover this procedure. The LE on the gPu in the standards used for campaign calibration purposes ranged from 0.4% to 1.9% for scrap standards and for waste standards was 3.2% or 3.5%. Most of this LE is due to the uncertainty

* This program, sponsored by the AEC, will, hopefully, introduce non-destructive measuring techniques in the routine handling of nuclear fuel thus providing more effective and timely material control within the plant.
LE CALCULATIONS USING DATA FROM NEUTRON
COINCIDENCE COUNTING

\[ y = aX + b \] general equation

From calibration data:

- \( a \) - slope
- \( b \) - intercept
- \( \sigma^2 \) - slope variance
- \( \sigma^2_b \) - intercept variance
- \( \text{cov}(a,b) \) - covariance between slope and intercept

\[ X_i = \frac{(y_i - b)}{a} \]

\[ X = \frac{\sum X_i = \sum (y_i - nb)}{a} \]

\( y_i \) - net corrected coincidence counts of sample \( i \).

\( X_i \) - calculated gPu 240 eq. cf sample \( i \).

\[ \sigma^2_X = \frac{\sigma^2(\Sigma y - nb)}{(\Sigma y - nb)^2} + \frac{\sigma^2_a}{a^2} - \frac{2 \text{ Cov}(\Sigma y - nb, a)}{(\Sigma y - nb)(a)} \]

\[ = \frac{\sigma^2_{\Sigma y} + n^2 \sigma^2_b}{(\Sigma y - nb)^2} + \frac{\sigma^2_a}{a^2} - \frac{2[\text{cov}(\Sigma y, a) - n \text{ cov}(b,a)]}{(\Sigma y - nb)(a)} \]

\[ \sigma^2_X = \left( \frac{\Sigma y - nb}{a} \right)^2 \left[ \frac{\sigma^2_{\Sigma y} + n^2 \sigma^2_b}{(\Sigma y - nb)^2} + \frac{\sigma^2_a}{a^2} + \frac{2n \text{ cov}(a,b)}{(\Sigma y - nb)(a)} \right] \]

If there was one calibration curve for the waste (measured discards) and \( J \) calibration periods for the scrap the limit of error expressions would be:

\[ \text{LE (waste)} = 2\sqrt{\sigma^2_X (\text{waste})} \]

\[ \text{LE (scrap)} = 2 \sqrt{\frac{\sum_{k=1}^{J} \sigma^2_X_k}{\sqrt{J}}} \]
associated with the plutonium concentration determination for the standards.

From the counting data collected on the known standards a calibration curve was fitted by using the least squares technique. The y axis, or ordinate, represented the coincidence counts for a fixed time corrected for accidental counts, dead time losses and background counts. The x axis, or abscissa, represented the grams of plutonium 240 equivalent in the standards. The weighting of calibration data had no significant effect on the resulting calibration parameters. From the least squares fit of the standards data the slope, intercept, standard deviations of the slope and intercept, and the covariance between the slope and intercept were obtained. The standard deviation of the slope and intercept represented the uncertainty associated with the standards provided that all the standards did not come from the same master blend. When the scrap and discarded materials were coincidence counted, the count, $\sum y_i$, was obtained with information for the counting variance. The following figure shows the pertinent expressions used to obtain the grams of plutonium 240 equivalent, $X$, and the associated variance, $\sigma^2_X$. Note that the summation of the individual gPu 240 equivalent values, $X_i$, takes place before the error propagation is applied. If this procedure is carried out in reverse order, the covariance effects among the individual counts would have to be accounted for. The $X_i$ values for all

-23-
of the material counted within a given calibration period were related through the use of the same calibration parameters. In other words, the variances associated with the individual $X_i$ values are not independent and cannot be summed to obtain the variance on the total, $\sigma^2_X$. On the other hand, variances from different periods based on independent calibrations can be summed together in the usual manner since they are not related. The procedure followed in the ISE is shown at the bottom of Figure 1.

A point that should be emphasized concerns the gPu 240 equivalent of the standards. If the standards used to generate a calibration curve all came from the same isotopic lot and master blend, the abscissa may be biased. This systematic error would not show up in the standard deviation of the intercept and slope values. It would then be necessary to account for this systematic error separately.

Because the neutron equipment was not stable over the three month period that the Campaign 1 waste and scrap material was counted it was not possible to use just one calibration curve. A study of the scrap standards calibration data showed that the system drifted upward in a quasi step function fashion resulting in four calibration periods for the three month time interval. A weighted regression analysis (linear least squares fit) was performed on scrap standards data for each of the four periods. Coincidence counting statistics were used as weights. Data analysis on the very weak waste
standards did not show any statistically significant trend. Therefore all neutron coincidence measurements on waste standards were used in one weighted regression.
Discussion on Campaign 2

Much of the above discussion on Campaign 1 applies to Campaign 2. The counting times were the same - 10 min. independent of whether the material was low level waste or recoverable scrap. Some of the standards used for calibration purposes were the same ones 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.

The problem of neutron counting equipment instability was present during the measurement of Campaign 2 waste and scrap. Therefore, there were several calibration curves and sets of parameters. The waste standards data and scrap standards data were not regressed separately by GE-VNC personnel within a given stable period as was done by NBS personnel in Campaign 1. This made the calculations within a given period simpler but caused complications when computing the LEMUF value because the waste and scrap values were not independent.

The problem of counting scrap packages which contained more plutonium than the standards was also present in Campaign 2.

For Campaign 2 regressions, weighting was done by GE-VNC on a mass-of-standard basis. An examination of the scrap standards data for a given period showed that the resulting intercept and slope values from least squares fits were
statistically the same when weighting was done by mass as when done by coincidence counting statistics.

Summary on Scrap and Waste

1. Campaign 2 measurement results were not expected to show improvement over those of Campaign 1 since the data collecting and data reducing procedures were the same for both campaigns.

2. On a percentage basis the scrap and waste uncertainties were larger for Campaign 2 because the plutonium in the standards came from different lots than was the plutonium used in Campaign 2. This created the situation where the impact of the isotopic abundance uncertainties as well as the spontaneous fission ratios uncertainties had to be evaluated and included in the limit of error calculations.
SUMMARY

Feed Material - PuO₂

The moisture problem which affected the plutonium dioxide of Campaign 1 was eliminated in Campaign 2 by equilibrating before sampling; this procedure permitted a meaningful chemical assay value to be obtained.

More effort on Pu 238 and Am 241 abundance determinations is needed if calorimetry is to do the job that this nondestructive technique is capable of performing for materials balance accounting.

Product Material - Sintered Mixed Oxide

The product % LE value, obtained from chemical assay, was the same for both campaigns, even though more blend preparation work was performed on Campaign 2 material.

There was a factor of approximately two between the % LE values of the two enrichments within each of the campaigns.

The variation in the % LE or %CV values between enrichments implies that this non-consistent process behavior will render nested design results useless for purposes of error propagation in material balance accounting, thus necessitating the requirement to obtain sufficient data for determining limits of error for each job.

Scrap and Measured Discards

The neutron coincidence counting procedure for measuring Campaign 2 materials was the same as that for Campaign 1.
The percent uncertainty on a given scrap item was higher in Campaign 2 because of the additional uncertainty due to the isotopic abundance and spontaneous fission ratio considerations. This additional complex uncertainty existed because the plutonium in the standards came from different lots than that used in Campaign 2.

**Overall Balance**

Campaign 2 was more successful on the whole than was Campaign 1, mainly as a result of the improved quantification of the feed material. The MUF and LEMUF values for Campaign 2 were in the 10 grams plutonium range as compared to a factor of 10+ larger for Campaign 1.
REFERENCES


