

# NON DESTRUCTIVE MEASUREMENT UNCERTAINTY AT A PLUTONIUM SCRAP RECOVERY FACILITY

**Joseph R. Wachter**  
Pajarito Scientific Corporation  
Los Alamos, NM 87544

Lynn A. Foster, Rudy J. Maez, John A. Pompeo  
Los Alamos National Laboratory  
Los Alamos, NM 87545

## ABSTRACT

---

Residues and wastes from a plutonium scrap recovery facility are often highly heterogeneous, span a wide range of chemical compositions and matrix types, and are packaged in a variety of container sizes. Because of this diversity, defensible estimates of bias and reproducibility in non destructive measurements of these materials are often problematic. An ongoing program for evaluating these uncertainty components over the broad spectrum of process residues and transuranic waste forms generated at the Los Alamos Plutonium Facility has been developed. This Measurement Verification Program has been in existence for more than 15 years and has revealed information on random and systematic uncertainties in the measurements that would be difficult to determine by other means. Two aspects of the program will be discussed in this report. First, original gamma-ray and neutron measurements will be compared with subsequent assays using the same technique. This comparison provides an estimate of the reproducibility in these measurements. Next, a comparison of segmented gamma scan and passive neutron coincidence measurements with values obtained by calorimetry and gamma-ray isotopics analyses will be conducted. This aspect of the program provides insights into measurement bias.

## INTRODUCTION

---

The Plutonium Facility at Los Alamos National Laboratory is a plutonium scrap recovery complex which is involved with chemical and metallurgical processing of a broad range of defense-related plutonium materials. Wastes and residues are routinely generated here from many plutonium metal and oxide processing steps. These processing steps produce a wide variety of leaner scrap and waste forms such as plutonium oxide from burned residues, Pu-bearing salts from production/reduction and metal purification processes, impure plutonium metal, metal reduction slags, ash, undissolved oxide heels, ceramics, cleaning rags, plastics, HEPA filters, and other remnants and apparatus generated from fabrication and cleanup tasks<sup>1</sup>.

Liquid wastes that result from plutonium processing include effluents from ion-exchange columns, oxalate filtrates, and caustic solutions generated by various head-end and purification operations. The solutions are filtered, treated in an evaporator, and chemically sampled to determine compliance with discard criterion. Nondestructive analyses (NDA) are not performed

on the liquid wastes. Upon meeting discard criterion, liquid wastes are fixed in cement and sent to a waste storage complex.

Solid residues and transuranic wastes (TRU) are first segregated according to matrix type. Then low bulk density materials such as paper and rags are assayed by Segmented Gamma Scanner analysis<sup>2</sup> (SGS); and higher density materials such as metals and most salts are measured with Passive Neutron Coincidence Counters<sup>3</sup> (PNCC). Plutonium isotopic identification is supplied by the generators. Prior to startup, all nondestructive assay instrumentation at this facility is put through a stringent series of qualification checks and measurements under expected operating conditions. Thereafter, instrument performance is monitored through daily and weekly stability tests, periodic calibration checks, and monthly, independent evaluation of instrument operation.

Despite instrument qualification assessments, measurement control procedures, and matrix segregation, nondestructive assays of the wastes and residues from plutonium processing operations are often troublesome and may pose formidable challenges to the measurement specialist. Reasons for the difficulties include:

- TRU waste and residues come in a variety of chemical compounds, physical sizes, isotopic proportions, and matrix compositions. Each of these may present complications for different measurement methodologies. Knowledge and quantification of these effects are difficult for many of the materials.
- At this facility, most of the residues and wastes are of a heterogeneous nature. That is, both the matrix and the plutonium are not evenly distributed throughout the container that is assayed. These spatial location variations can limit assay reliability.
- Representative standards for many kinds of residues and wastes do not exist and may prove impossible to fabricate. Indeed, the heterogeneous nature of many of these materials defy all attempts to match them with representative standards.

In order to maintain additional confidence in the assays performed at the Los Alamos Plutonium Facility, studies are routinely conducted to monitor the reliability of NDA measurements of plutonium-contaminated residues and waste. One of these studies is an ongoing program for verifying measurements over the broad spectrum of matrix forms and plutonium loadings that are generated here. This Measurement Verification Program (MVP) was originally intended as a mechanism for resolving inventory differences, verifying the nuclear material content of items, and identifying problem measurement materials. But the data collected over the 15 year span of the program have also revealed information on random and systematic uncertainties in the measurements that would be difficult to determine by other means. Although the program generally focuses on residues, that is, on materials that have higher concentrations of plutonium than most waste forms; their chemical composition, matrix, instrument selection, and packaging are equivalent to most TRU waste generated here. Therefore, these examinations of instrument performance for residues can be extended to provide insight into the performance of the instruments for measuring waste.

There are several segments to the program, however only the two that pertain to leaner residues and waste will be included in this report. One pertinent segment compares SGS and PNCC analyses with previous measurements performed by the same technique. For example, items that have been measured initially by an SGS will receive a second measurement at a later time with either the same instrument or a different SGS. This comparison gives an indication of the measurement reproducibility that can be expected from a given technique. A second segment

compares SGS and PNCC measurements with reference values obtained from calorimetry + gamma-ray isotopics (CI) analysis. These comparisons are useful in determining measurement bias associated with NDA techniques. A discussion of the results obtained from these segments of the program is given below.

## MEASUREMENT VERIFICATION PROGRAM

---

The MVP is applied to the entire range of NDA instruments that are used to measure residues and TRU waste. Process materials containing plutonium as an oxide, a salt or metal; and embedded in a wide variety of matrices typical of a scrap recovery facility, are assayed with these instruments. That is, rags, tools, crucible parts, plastics, rubber and leaded gloves, furnace parts, non plutonium metal, etc. are all analyzed with these instruments. A total of six SGSs and eleven PNCCs have been used since the inception of the program. These instruments have measured over 95 % of the leaner residues and wastes generated at this facility. Plutonium content varies according to chemical and matrix type. However, for the purposes of this study, the comparison range for SGSs has been restricted to between 1 and 220 grams of plutonium, and to between 1 and 240 grams for PNCCs. The assays for both segments of the program are single measurements of 20 minutes, or less, duration. During the course of the program, all instruments were under the measurement control procedures cited above. For that segment of the program in which items are assayed by the same technique, measurement intervals ranged from one week to 13 years. On average, however, the two measurements were separated by about a three year interval. Providing the instruments used for the assays have been operating in a stable fashion over the time period separating the measurements, as guaranteed by the measurement control procedures, this comparison gives an estimate of the long term reproducibility of the measurement for the NDA technique and type of process material under review. For the segment of the MVP which compares SGS or PNCC assays to reference CI analyses, the two measurements are generally separated by about four years. Because previous studies<sup>4</sup> have indicated that the reference measurements are accurate to within  $\pm 1\%$  of the nominal plutonium value in the sample, this information provides a gauge to estimate measurement bias.

Table 1 summarizes the results from these two segments of the MVP data. Ratio  $R_1$  in the table is the average of the ratios of a single SGS or PNCC measurement to a second measurement using the same technique made at a later date. The ratio  $R_2$  is the average of the ratios of a single SGS or PNCC measurement to that of a reference CI measurement. Results from all SGSs and PNCCs were folded together to obtain the reported ratios. No weighting of the data to account for plutonium mass loading was performed. The uncertainties associated with the ratios are the standard deviations (1s) of the combined data in each category. Some data were removed from the database prior to calculation of the ratios. Four reasons were used to justify their removal. First, all measurements made prior to 1982 were deleted because numerous anomalies occurred in this data set and because less rigorous measurement control mechanisms were in use before that time. Second, assay data that were outside of instruments' calibration ranges were not included here. Third, measurements that were performed on instruments that later proved to be inappropriate for that matrix type were taken off the database. During the course of the MVP program, it was noted that three matrix types have been assayed with an inappropriate technique. Because of this, ad hoc studies were developed to determine the best measurement strategy for those matrix types. MVP data taken before the new strategies were implemented have therefore been removed from this study so that their bias would not be reflected in the results. Fourth, measurement data were deleted during periods when an instrument's measurement control program indicated that the instrument was operating outside of control limits.

	<b>R<sub>1</sub></b>	<b>R<sub>2</sub></b>
<b>SGS</b>	1.04 ± 0.20 (427)	1.01 ± 0.16 (63)
<b>PNCC</b>	1.00 ± 0.13 (348)	1.03 ± 0.13 (125)

**Table 1:** Results from analyses of all SGS and PNCC MVP data are summarized below. R<sub>1</sub> is the average of the ratios of a single SGS or PNCC measurement to a second measurement made at a later time using the same NDA technique. R<sub>2</sub> is the average of the ratios of SGS or PNCC measurements to CI results. The numbers in parenthesis are the number of measurements used to determine each of the ratios.

The table indicates that R<sub>1</sub> is 1.04 for SGSs; that is, later SGS remeasurements averaged 4 % lower than the initial measurements. This ratio is consistent with settling of heavier plutonium particles inside the containers between the times the two measurements were performed. Settling has been observed to occur for some of the salts measured by this technique. When denser plutonium particles fall to the bottom of the container between the earlier and later measurements, the resulting self attenuation and end effect counting losses tend to bias the later assay low. The ratio R<sub>2</sub> for SGSs is 1.01, indicating that these measurements are slightly larger, on average, than the reference measurements. However, the magnitude of this bias (1%) is small compared to the standard deviation in the data (16%), so conclusions on bias for these measurements are problematic. For PNCCs, R<sub>1</sub> equals 1.00, indicating no difference, on average, between initial and later measurements using this technique. The ratio R<sub>2</sub> for PNCCs is 1.03, indicative of an average high bias of 3% in these measurements. This result may reflect uncorrected multiplication and (a,n) effects which tend to overestimate neutron counter assays. Again, the magnitude of the bias is small compared to the standard deviation in the data. Figures 1-4 depict graphically the results of these four measurement studies.

Several additional comments can be made with regard to the results of Table 1. First, each SGS in the study exhibits similar reproducibility tendencies. That is, for each of the SGSs used in the study, the later measurements are consistently smaller, on average, than the earlier measurements. Thus, in addition to the overall SGS R<sub>1</sub> average being greater than unity, the same is true for each of the individual SGSs. A second important comment concerns the large standard deviations for each of the program segments reported in Table 1. These large variabilities are not isolated to just a few matrix types, but instead are spread across the entire spectrum of materials and instruments that were included in the program. Large bias variabilities may be due, in part, to uncertainties in the plutonium isotopic values used in the SGS and PNCC assays. As indicated above, isotopic identification is supplied by generators of the residues and waste. They generally reflect broad averages defined from plutonium product streams that receive destructive analysis. While these averages have proven correct for the entirety of product, residue, and waste materials generated here, they may be inadequate for the individual residue and waste streams included in the MVP program. In addition, these large variabilities are also indicative that there are some matrices for which no NDA technique is entirely suitable. Certain salts generated at this facility fit this category.

[INSERT FIGURES 1-4]

The MVP data were also subdivided according to matrix type. These subdivisions, shown in Tables 2 through 5, indicate reproducibility and bias for most of the materials included in the MVP program. The ratios shown in the tables include all instruments used to assay that matrix category. Again, the data have been filtered to remove measurements of inappropriate matrices

and items that were assayed outside instrument calibration ranges. Because of this filtering, no apparent biases or indications of instrument instability can be discerned for any of the matrices. Again, large standard deviations in the data ( $\approx 8\%$ ) are common. The reproducibility ratio  $R_1$  varies from a low of 0.94 (Electrorefining Salts) to a high of 1.12 (Sand, Slag, and Crucible) for SGSs and from 0.95 (Rags) to 1.08 (HEPA filters) for PNCCs. Variation in the bias ratio  $R_2$  ranges from 0.91 (Miscellaneous Residues) to 1.06 (Hydroxide Cake) for SGSs and from 0.88 (Sweepings) to 1.10 (Electrorefining Salts) for PNCCs.

<b>Matrix</b>	<b><math>R_1</math></b>
Hydroxide Cake	$1.03 \pm 0.09$
Electrorefining Salts	$0.94 \pm 0.15$
HEPA Filters	$1.11 \pm 0.28$
Leached Solids & Fines	$1.03 \pm 0.15$
Slag & Crucible from Fluoride Reduction	$1.02 \pm 0.10$
Plastics, Paper, Rags	$1.02 \pm 0.19$
PuO <sub>2</sub> Scrap from Various Operations	$1.00 \pm 0.13$
Miscellaneous Residues	$1.10 \pm 0.27$
Salts Residues	$0.97 \pm 0.13$
Sand, Slag, & Crucible	$1.12 \pm 0.29$

*Table 2: SGS reproducibility results listed for matrix categories included in the MVP program.*

<b>Matrix</b>	<b><math>R_2</math></b>
Hydroxide Cake	$1.06 \pm 0.17$
Electrorefining Salts	Insufficient Data
Leached Solids & Fines	$0.95 \pm 0.09$
PuO <sub>2</sub> Scrap from Various Operations	$1.05 \pm 0.17$
Miscellaneous Residues	$0.91 \pm 0.11$
Sand, Slag, & Crucible	$1.04 \pm 0.10$

*Table 3: SGS bias as a function of matrix type.*

Matrix	R <sub>1</sub>
Hydroxide Cake	1.00 ± 0.15
Electrorefining Salts	1.00 ± 0.08
HEPA Filters	1.08 ± 0.20
Non Pu Metals	1.03 ± 0.17
Slag & Crucible from Fluoride Reduction	1.04 ± 0.12
PuO <sub>2</sub> Scrap from Various Operations	0.98 ± 0.12
Rags	0.95 ± 0.12
Miscellaneous Residues	0.96 ± 0.12
Salt Residues	1.05 ± 0.16
Sweepings	1.02 ± 0.08

**Table 4:** PNCC reproducibility for matrix categories included in the MVP program.

Matrix	R <sub>2</sub>
Hydroxide Cake	1.08 ± 0.18
Electrorefining Salts	1.10 ± 0.10
Non Pu Metal	Insufficient Data
PuO <sub>2</sub> Scrap from Various Operations	0.90 ± 0.17
Miscellaneous Residues	1.00 ± 0.09
Salt Residues	1.02 ± 0.09
Sweepings	0.88 ± 0.13

**Table 5:** PNCC bias as a function of matrix type.

An important consideration for the comparisons made in the MVP program is the extent to which the program's data can be applied as valid uncertainty estimators to residue and TRU waste measurements. Because uncertainty elements in the MVP measurements (such as matrix variations, plutonium chemistry, container sizes, heterogenities, etc), are also the major components in measurement error for residues and wastes, an argument can be made that they are valid estimators. This is true for each of the NDA techniques and matrix categories that have been included in the program. In defense of this argument, several additional programs that independently assess measurement performance have not identified problems during the last seven years. These additional programs include ad hoc studies of potentially problematic materials and real time inventory difference evaluation for most plutonium processing streams. However, other aspects of the program may limit its applicability. One of these aspects is that the MVP often focuses on problem materials which are not always representative of overall residue and waste throughput at this facility. This focus would tend to overestimate measurement bias because biases tend to be higher for problem materials. Also, the majority of MVP measurements are performed on residues and only a fraction are performed on wastes. This also may limit the program's application and comes about, in part, because TRU wastes cannot be stored at this facility for the lengthy intervals between MVP measurements; and, in part, because reference calorimetry analyses cannot be performed on many waste containers due to their size and low plutonium loadings. A final limitation in the program is that it may take years to develop an extensive database that identifies measurement uncertainty over the entire range of residue and waste categories that are typical of a plutonium scrap recovery facility. Here, the difficulty takes place because these facilities produce residues and waste in a wide variety of matrix forms, container sizes plutonium mass loadings, heterogenities, etc. and because reference values may

be difficult to ascertain. These both tend to lead to the requirement for a significant measurement effort to maintain the program and for long program development times.

## **SUMMATION**

---

Reported above are the results of a program to estimate reproducibility and bias in measurements of residues and wastes at the Los Alamos Plutonium Facility. SGS measurements of these materials were found to be reproducible to within 4%, on average, and to be subject to an average bias of 1% over the broad range of materials included in the program. Passive neutron counter measurements were reproducible to within 0.5% and had a bias of 3% when compared to reference values. Large variabilities were noted in all bias and reproducibility data in the program.

## REFERENCES

---

1. D.C. Christensen, B.F. Bowersox, B.J. McKerley, and R.L. Nance, *Wastes from Plutonium Conversion and Scrap Recovery Operation*, Los Alamos National Laboratory Report LA 11069, Los Alamos, NM, 1988.
2. E.R. Martin, D.F. Jones, and J.L. Parker, *Gamma-Ray Measurements with the Segmented Gamma Scan*, Los Alamos National Laboratory Report LA 7059-M, Los Alamos, NM, 1977.
3. M.S. Krick and H.O. Menlove, *The High-Level Neutron Coincidence Counter (HLNCC) User's Manual*, Los Alamos National Laboratory Report LA 7779, Los Alamos, NM June 1979.
4. V.L. Longmire, T.L. Cremers, W.A. Sedlacek, S.M. Long, A.M. Scarborough, and J.R. Hurd, *Isotopic Ratios and Effective Power Determined by Gamma-Ray Spectroscopy vs Mass Spectroscopy for Molten Salt Extraction Residues*, Proc. of 31<sup>st</sup> Annual Institute of Nuclear Materials Management Meeting, Los Angeles, 1990.