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NON-DESTRUCTIVE MEASUREMENT OF SOLID PLUTONIUM WASTE AT LOS ALAMOS NATIONAL LABORATORY

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INTRODUCTION

Los Alamos National Laboratory (LANL) is a national defense facility involved in the recovery and processing of plutonium. Wastes and residues are routinely generated here from many stages of plutonium metal fabrication and from pyrochemical and aqueous processing of plutonium scrap. Materials which require measurement include 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, and auxiliary implements such as HEPA filters, plastics, and cleaning rags.[1] Nondestructive assays (NDA) of transuranic (TRU) waste from these materials are often troublesome and may pose formidable challenges to the measurement specialist. Reasons for difficulties include:

- TRU waste comes in a variety of chemical compounds, physical sizes, isotopic proportions, and matrices. Each of these may present complications for different instrument methodologies.

- Many forms of TRU waste are of heterogeneous composition. The presence of metal shot and variations in matrix densities can cause attenuation problems in gamma-ray and x-ray based instruments and multiplication and (α,n)

effects in neutron counters.

- TRU waste is often packaged in large receptacles containing small amounts of plutonium. Thus, they are often not amenable to highly accurate measurement techniques such as calorimetry or spectrochemical analyses.

- Representative standards for many kinds of waste materials do not exist yet and may prove difficult or impossible to fabricate. Indeed, the nature of many heterogeneous waste samples defy attempts to match them with representative standards.

Various NDA strategies are employed to assay plutonium waste. These incorporate gamma-ray and x-ray based measurement techniques, passive and active neutron counting, and to a lesser extent, calorimetry. All of these can provide reasonably good analyses for certain categories of waste materials, but suffer from deficencies if applied to categories having unsuitable matrices, plutonium mass ranges, chemical compositions, etc. This report will briefly present the NDA strategies employed at LANL for measurement of solid wastes and residues which do not require immobilization, then review studies performed here on the reliability of NDA measurements of TRU waste. Emphasis will be placed on assays

of Pu-bearing salts. These materials are generated from several pyrochemical preparation and purification processes including plutonium tetrafluoride production/reduction, molten salt extraction and direct oxide reduction. The salts may contain unknown quantities of metal shot, high americium content, and be mixed with fluoride, all of which can pose impediments to reliable NDA analysis. Following the salt studies, the report will briefly discuss the susceptibility of passive thermal neutron drum counters to matrix moderating effects. The matrices are representative of the major waste categories produced at the LANL Plutonium Facility. Finally, some closing remarks will include a short discussion of several unresolved waste measurement issues here.

LANL WASTE MEASUREMENT STRATEGIES

The first step in the waste measurement process at Los Alamos is the proper segregation of materials according to chemical structure, physical size, isotopic composition, and matrix content. Waste measurements of paper, plastics, cellulosics, ash, powders, and other low density materials contaminated with plutonium are made with Segmented Gamma Scanners (SGS). The SGSs used here are capable of making measurements of a broad array of container sizes, including 208 liter drums. These instruments use the gamma rays from an external transmission source to develop correction factors for

attenuation in the sample and container. The factors are applied to a segmented, high resolution, gamma-ray measurement of the plutonium signal to determine the assay value. Providing the waste matrix is reasonably uniform and self-attenuation of gamma-rays within the plutonium particles can be neglected, this technique provides a reliable method for assaying waste ranging in plutonium content from a few grams up to several hundred grams. Measurement accuracies of 5% to 10%[2] for many waste materials can be achieved using measurement times of less than 15 minutes.

Measurements of high density plutonium waste such as metals, leaded gloves, tools, motors, etc., and some intermediate density salt residues are made using Thermal Neutron Counters (TNC) at LANL. The facility has TNC well counters in several sizes for smaller containers and a large counter for samples as large as 208 liter drums. These instruments monitor the correlated neutrons from the spontaneously fissioning isotopes of plutonium to produce an assay. If moderation and absorption effects in the matrix are minor and if neutron multiplication in the sample is small, these instruments also produce reliable measurements of plutonium waste ranging from a few grams up to several hundred grams. Accuracies of 5% to 10%[3] can be realized for many waste items with measurement times of less than 15 minutes.

Calorimetry is an NDA technique for measuring the heat associated with the decay of radioactive materials. Much of the decay energy of plutonium is released as alpha or beta particles and converted to heat inside the sample. Calorimetric assay consists of a calorimetry measurement of the heat output from plutonium coupled to a gamma-ray isotopic analysis of the sample specific power (the sum of the products of the measured isotopic fractions and the known heat output per gram of each isotope). Several isotopic codes are presently used here. A combined calorimetry/isotopics (C/I) analysis produces highly accurate and repeatable assays; random and systematic errors of less than 1.5% (1σ) can be achieved for many categories of plutonium-bearing materials. However, because of the low heat output and large volume of TRU waste, and because of the long assay times required by C/I analysis, these instruments are not generally used to assay waste materials.

To assure confidence in the assays performed at LANL, studies are routinely conducted to monitor the reliability of the NDA measurements of the plutonium products, residues, and wastes generated here. The studies frequently focus on materials that resist simple measurement methodologies such as pyrochemical salts and scrap. The next four sections of this report will discuss studies which have been carried out here

in the recent past.

HYDROFLUORINATION WASTES

These materials consist of product slag mixed with magnesium sand and crucible remnants resulting from PuF_4 production and reduction. The slag, containing CaF_2 salt and unknown amounts of PuF_4, may also be mixed with significant quantities of plutonium metal shot. This matrix presents difficulties for TNC analysis because of large (α,n) effects resulting from the presence of fluorine atoms. Likewise SGS assays are hindered by attenuation of the plutonium gamma rays in the shot. A study was performed to determine the character of the biases resulting from both measurement methods. The study compared multiple SGS and TNC measurements of hydrofluorination wastes to C/I analysis. Because of the accuracy of C/I measurements, these were chosen as the reference measurements. The plutonium content in the wastes extended from 10 g to 300 g. Results of the assays are summarized in Table I. They indicate that nearly all the SGS measurements in this range were biased low. Although a bias was expected, there was no predictable pattern to the differences between the SGS and reference measurements. While some of the SGS measurements were within a few grams of the C/I values, others disagreed by more than 50% of the reference value. The unpredictability of the differences, thought to be due to

differences in self-attenuation resulting from variations in shot size, makes determination of SGS bias correction factors difficult.

A comparison of the TNC measurements with those by C/I indicated a bias in the opposite direction. The presence of PuF_4 in the slag would again lead one to expect the bias. In this case however, the pattern of the bias was more predictable since 95% of the TNC measurements were between 8% to 26% higher than the C/I results. Because the TNC results predictably differed from the reference measurements, a bias correction factor was calculated for TNC analysis of these materials. A weighted least squares fit of the TNC data for plutonium-bearing items between 0 and 200 grams* indicated that,

$$M(TNC) = KM(C/I)$$

where M is the plutonium mass determined either by TNC or C/I analysis and K is the bias correction factor.

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* Calorimetry/Isotopic assays are generally used to measure items containing more than 200 grams of plutonium because of the decreased sensitivity of TNCs for quantities above this amount and because of more stringent accountability requirements for the larger items. Hydrofluorination materials with higher plutonium loadings are measured here using C/I analysis.

Application of this simple, linear correction to the TNC assay result provided an improved measurement strategy for wastes generated by this process. These can now be quickly and reliably measured without resort to time-consuming analysis by destructive means or C/I. Figure 1 shows the agreement between TNC and C/I measurements after the correction has been applied. Only two of the TNC measurements fall outside ±10% of the C/I values, indicating reasonable concurrence between the two results for these difficult materials. The correction factor is currently applied to assays of hydrofluorination slags at LANL and the validity of the factor is checked periodically by further comparisons with C/I measurements.

MOLTEN SALT EXTRACTION WASTES

Purification of impure plutonium metal is often performed by Molten Salt Extraction (MSE) to reduce the ^{241}Am content in the metal product. A byproduct of this purification procedure is a salt mixture consisting of KCl, NaCl, MgCl, $PuCl_3$, and $AmCl_3$ and often containing shards of MgO crucible.[3] The salts may contain up to several grams of americium which can hinder TNC measurements due to (α,n)- induced fissions and to count rates too large for conventional preamplifiers to process. SGS assays are also impeded on account of the presence of plutonium metal slot. Because of these difficulties, a study was performed to evaluate the effectiveness of TNC, SGS, and

C/I measurements. In the study, averaged values from multiple TNC, SGS, and C/I measurements were compared to reference values obtained from Isotopic Dilution Mass Spectroscopy (IDMS) performed by the LANL analytical chemistry group. Material samples were selected which ranged in plutonium content from 56 g to 409 g. Two SGS measurement algorithms were used in the study; one used the conventional sample attenuation correction mentioned previously while the other used both the conventional correction plus a prototype lump-correction algorithm developed at LANL.[4] TNC measurements[5] were made with improved preamplifier electronics which could process the high count rates characteristic of MSE wastes, however no corrections based on (α,n) content or neutron multiplication were applied to the data. The results, shown in Table II, indicate that biases resulted from all three measurement techniques. The presence of metal shot limits both conventional and lump-corrected SGS measurements. Each was biased low by 4%, on average, compared to the reference values, however there was less variability in the lump-corrected measurements. Due to (α,n) effects in these materials, the TNC measurements were uniformly high. The bias ranged from a minimum of 7% to a maximum of 120% of the IDMS values, and had a tendency to increase for samples with larger quantities of plutonium. Finally, the C/I measurements also showed a high bias which averaged 4% for this study. While

the causes of this bias are not fully understood, it was noted that the isotopics analysis consistently underestimated the ^{241}Am fraction which in turn caused the plutonium content to be overstated. Additional studies comparing several isotopic measurement algorithms are currently underway to more fully quantify the C/I bias.

Due to the difficulties in assaying these materials by conventional NDA methods, current practice here is to perform SGS measurements only on items containing less than 50 grams of plutonium (no SGS bias has been determined for low Pu loadings); larger samples require calorimetry and isotopics analyses. Because of poor heat conductivity in these materials, long calorimetry measurement times[3] (up to 24 hrs) are required. This, in turn, results in decreased measurement throughput at this facility. These materials remain one of the most difficult NDA measurement challenges encountered here.

DIRECT OXIDE REDUCTION SALTS

In Direct Oxide Reduction (DOR), plutonium oxide is reacted with a charge of calcium metal in a $CaCl_2$ (or $CaCl_2$ 20 mol% CaF_2)[1] solvent to produce plutonium metal and calcium oxide. On completion of the reaction, the spent salt, containing $CaCl_2$, CaF_2 and low levels of ^{241}Am, is provided with an NDA

measurement to determine the plutonium content. Typically salts from this process are too dense to allow SGS analysis, and TNC measurements have been used for their assay. However, the presence of fluoride makes TNC assays questionable and necessitated a study to compare the results of TNC and reference IDMS measurements. In the study, multiple TNC measurements of DOR salts ranging in plutonium content from 100 g to 470 g were averaged and compared to IDMS values obtained from blended samples. The TNC values were found to be larger than the reference values by 3.5%, on average. As the uncertainty associated with the TNC measurements, here defined as the relative standard deviation of the multiple measurements, was typically on the order of 6.5% for DOR residues, the systematic error was less than the random error of the measurements, making analysis of the comparison more difficult. At present, additional studies involving improved neutron counting techniques[6] are being investigated for measurements of waste generated from this process. Current measurement practice at LANL entails the usage of TNCs for plutonium loadings up to 250 g, and C/I analysis for higher loadings.

MODERATION EFFECTS IN TNC DRUM COUNTERS

Crucial to the acceptance of passive TNC measurements of TRU waste is an understanding of matrix moderating effects in

these instruments. Knowledge of these effects is particularly important for wastes because waste is often packaged in 208 liter drums which contain large quantities of matrix constituents. Corrections for matrix effects are incorporated into SGS measurement algorithms through the attenuation correction which primarily reflects effects due to the low density matrix. However, for waste materials not amenable to SGS analysis, passive TNC measurements are generally employed here. To determine the susceptibility of TNC drum counters to matrix variations, a study[7] was performed to determine the effects due to various matrix categories. For the study, materials representing the major solid waste matrix categories (but not including immobolized waste) generated at the LANL Plutonium Facility were simulated, loaded into 208 liter drums, then assayed together with PuO_2 standards. The measurement results were then compared to the nominal standards' values to determine if differences resulted from variations in the matrix materials. The waste categories were simulated as follows:

- Metals and high density matrices were simulated by distributing 35.0 Kg of scrap steel evenly about the plutonium standards.

- Salt, slag, crucible, and medium density matrices were

imitated by evenly distributing first, 9.0 Kg and second, 18.0 Kg of SiO_2 about the standards.

- Cellulosics and low density matrices were simulated first, by surrounding the standards with 8.0 Kg of plastic (vinyl bakelite) and second, with 1.8 Kg of cheese cloth.

The plutonium content of the standards ranged from 45 g to 179 g of (total) plutonium. The data were corrected for background, instrument deadtime, container moderation effects, and instrument height detection efficency. The results are summarized in Figure 2 for each of the matrices simulated. The figure graphs the differences between the measured and nominal mass values of the standards to the nominal mass value (determined by chemical analysis). In summary, the results indicate that:

1) No bias resulting from neutron moderation is present for cloth, SiO_2 (9 Kg or 18 Kg), or metal.

2) A small, high bias may exist for the plastic matrix. The average bias of the four points is 2.85% and comes about from the large hydrogen content in this material.

As a result of the study's findings, the existing practice of using this instrument to measure plutonium waste primarily in high and medium density matrices was vindicated. Plastics and other low density matrices represent only a small fraction of the throughput of the TNC drum counter and a correction factor is applied when appropriate.

UNRESOLVED WASTE MEASUREMENT ISSUES AT LANL

Many factors can affect the integrity of NDA waste measurements. The factors are often subtle and may be outside the assayer's control or knowledge. At LANL some of the factors that have an ongoing effect on NDA assays are listed below:

- Changes in Instrument Response: Vigilance must be constantly maintained for effects resulting from RF noise, variations in electrical power, or deterioration of detector response with time. The effects may be difficult to detect; for example, cumulative neutron damage to gamma-ray detectors or seasonal variations in the baseline voltages of calorimeters are difficult to observe but, on several occasions, have materially affected C/I measurements here.

- Modifications in TRU Processing: To improve processing efficiency, TRU producers and reprocessors will fre-

quently modify production steps, alter processing chemistry, or otherwise change the form or composition of TRU waste. These changes can lead to unexpected difficulties in the measurement of radioactive materials. At LANL, this has led at different times to high concentrations of metal shot in some salts, increased ^{241}Am concentrations in some wastes, altered TRU/salt matrix ratios, and increased bulk densities in TRU waste. All of these changes affected the NDA results and all were initiated without warning the people responsible for performing the assays. Because NDA measurements can be affected, it is imperative that proposals for production changes be promptly communicated to waste measurement personnel.

- Packaging Effects: In many TRU production and processing facilities, standardization of containers is not practiced. Variations in the size, shape, composition, or thickness of TRU containers can lead to measurable effects in waste assays. Here, changes in containers have led to altered SGS assays because no container attenuation factor was available for the new can. TNC assays have also been affected when samples were surrounded with unexpected amounts of highly-moderating packaging materials.

- Matrix Effects: Even in situations where NDA technologists believe they are cognizant of all matrix effects in assays of TRU materials, vigilance must be maintained. At LANL, we have noticed that Pu/salt matrix concentrations, ^{241}Am content, and plutonium shot quantities may vary dramatically in a given process depending on the stage of completeness of a chemical reaction. Thus for example, incomplete processing has resulted in unexpected amounts of salt or shot in residues which in turn affected the reliability of the NDA assay.

- Changes in Calibration Standards: Settling can occur in both homogeneous and "representative" heterogeneous standards. If homogeneous standards mixed with diluents are not regularly shaken, the plutonium may migrate to the bottom of the container, thereby affecting instrument calibration. Heterogeneous standards pose more challenging difficulties. If the standards were fabricated with lumps of plutonium oxide or salt to simulate process material, their composition may change with time due to partial pulverization of the lumps when the standard is handled. The resultant redistribution of plutonium inside the container can be particularly harmful to C/I and SGS calibrations. Settling in both

kinds of standards has been noted here on several occasions.

These factors, and many others left unmentioned, have each temporarily impaired NDA assays of TRU waste at LANL. To meet these measurement challenges, surveillance and evaluation programs have been implemented to scrutinize assays of materials from many waste streams generated in this facility. Some emerging NDA technologies, such as lump-corrected SGS[4] improved neutron self-interrogation techniques[6], moisture content corrections in neutron counters[8], flattened energy and spatial response profiles in active and passive neutron counters[9], and new techniques for measuring (α,n) and multiplication effects[10,11], may alleviate some of these problems. However, the usefulness of these technologies for measuring highly heterogeneous TRU waste, containing indeterminant and mixed matrices, TRU lumps, packaging variations, etc. remains unproven. Until such time as they are proven, constant monitoring and evaluation of the instruments and measurements is the only means to ensure the reliability of NDA waste assays.

REFERENCES

1. D. C. Christensen, D. F. Bowersox, B. J. McKerley, R. L. Nance, "Wastes from Plutonium Conversion and Scrap Recovery Operations", Los Alamos National Laboratory Report, LA-11069 (1988).

2. F. J. Schultz and J. T. Caldwell, "DOE Assay Methods Used for Certification of Contact-Handled Transuranic Waste", Oak Ridge National Laboratory Report #6485 (1989).

3. V. L. Longmire, W. E. Sedlacek, J. R. Hurd, A. M. Scarborough, "A Comparison of Conventional and Prototye Nondestructive Measurements on Molten Salt Extraction Residues", Proceedings of the Third International Conference on Facility Operations - Safeguards Interface, San Diego, CA, Nov. 29 - Dec. 4, 1987.

4. J. K. Sprinkle, Jr. and S. T. Hsue, "Recent Advances in Segmented Gamma Scanner Analysis", Proceedings of the Third International Conference on Facility Operations - Safeguards Interface, San Diego, CA, Nov. 29-Dec. 4, 1987.

5. J. K. Sprinkle, Jr., G. E. Bosler, S. T., Hsue, M. P. Kellogg, M. C. Miller, S. M. Simmonds, and A. R. Smith, "Nondestructive Assay of Plutonium-Bearing Scrap and Waste", to be presented at the Remote Systems Technology Division/American Nuclear Society meeting, San Francisco, CA, Nov. 26 -30, 1989.

6. P. A. Russo, H. O. Menlove, K. W. Fife, M. H. West, and B. L. Miller, "Evaluation of the Neutron Self-Interrogation Approach for Assay of Plutonium in High (α,n) Materials", Proceedings of the Third International Conference on Facility Operations - Safeguards Interface, San Diego, CA, Nov. 29 - Dec. 4, 1987.

7. J. R. Wachter, A. M. Scarborough, Los Alamos National Laboratory, unpublished data.

8. J. E. Stewart and H. O. Menlove, "Moisture Corrections in Neutron Coincidence Counting of PuO_2", Proceedings at the Third International Conference on Facility Operations - Safeguards Interface, San Diego, Ca, Nov. 29-Dec. 4, 1987.

9. H. O. Menlove, R. Palmer, G. W. Eccleston, N. Ensslin, "Flat-Squared Counter Design and Operation Manual", Los Alamos National Laboratory Report, LA-11635 (1989).

10. M. S. Krick and J. E. Swansen, "Neutron Multiplicity and Multiplication Measurements", Nucl. Inst. and Meth. 219, p. 384 (1984).

11. J. R. Wachter, E. L. Adams, N. Ensslin, "Prototype Fast Neutron Counter for the Assay of Impure Plutonium", Proceedings of the Third International Conference on Facility Operations - Safeguards Interface, San Diego, CA, Nov. 29 - Dec. 4, 1987.

Table I. Summary of the SGS, TNC, and C/I measurements of hydrofluorination wastes. The C/I results are presented as the reference measurements and the SGS and TNC values are ratioed to them.

ITEM	C/I (g Pu)	SGS C/I	TNC C/I
1	35		1.23
2	54		1.26
3	73		1.15
4	33	0.94	1.40
5	64	0.73	1.16
6	134	0.50	1.22
7	156	0.55	1.26
8	192	0.68	1.22
9	140	1.04	1.19
10	185	0.92	1.13
11	223	0.60	1.09
12	300	0.87	1.11
13	124	0.73	1.21
14	294	0.38	
15	127	0.94	
16	132	0.94	
17	166	0.88	
18	199	0.79	
19	136	0.95	
20	201	0.95	
21	216	0.92	
22	16		1.08
23	107	0.70	1.18
24	192	0.68	1.22
25	27		1.22
26	10		1.20
27	27		1.26

Table II. This table lists the reference IDMS values and the ratios of conventional SGS, lump-corrected SGS, TNC, and C/I measurements to the reference values. The averages of the ratios are given below each column.

ITEM	IDMS (g Pu)	Conventional SGS / IDMS	Lump-Corr[4] SGS / IDMS	TNC / IDMS	C/I / IDMS
1	55.56	1.01	-	-	1.03
2	90.39	1.00	0.95	1.35	1.03
3	141.21	0.91	-	-	0.99
4	155.37	0.97	0.96	1.54	1.09
5	243.83	0.98	0.95	1.95	1.04
6	246.95	0.78	0.96	1.07	1.01
7	263.56	1.07	0.97	1.72	1.05
8	372.73	0.98	0.95	2.12	1.10
9	408.58	0.92	0.97	2.20	1.06
	AVG	0.96	0.96	1.71	1.04

FIGURE CAPTIONS

Figure 1. The data points compare the results of measuring hydrofluorination wastes by bias-corrected TNC and C/I. The straight line is a weighted least squares fit of the TNC data.

Figure 2. The figure indicates moderation effects in the TNC drum counter by graphing the difference between the measured and nominal values of the standards as a function of the nominal mass value. The relative uncertainty due to random error for each data point is approximately 2.5%.

HYDROFLUORINATION WASTES
Corrected TNC Data

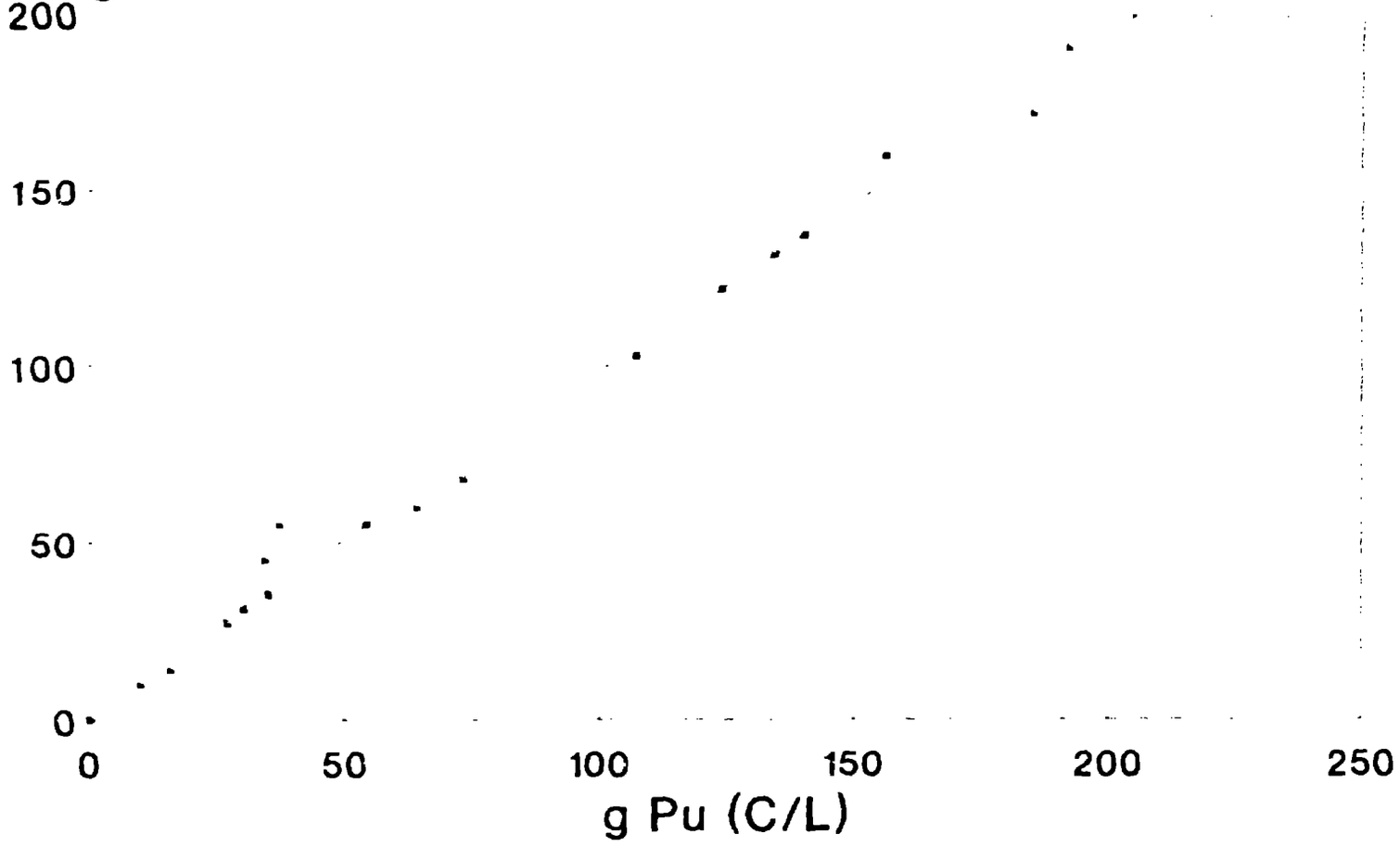

Figure 1

MODERATION EFFECTS IN TNC DRUM COUNTER

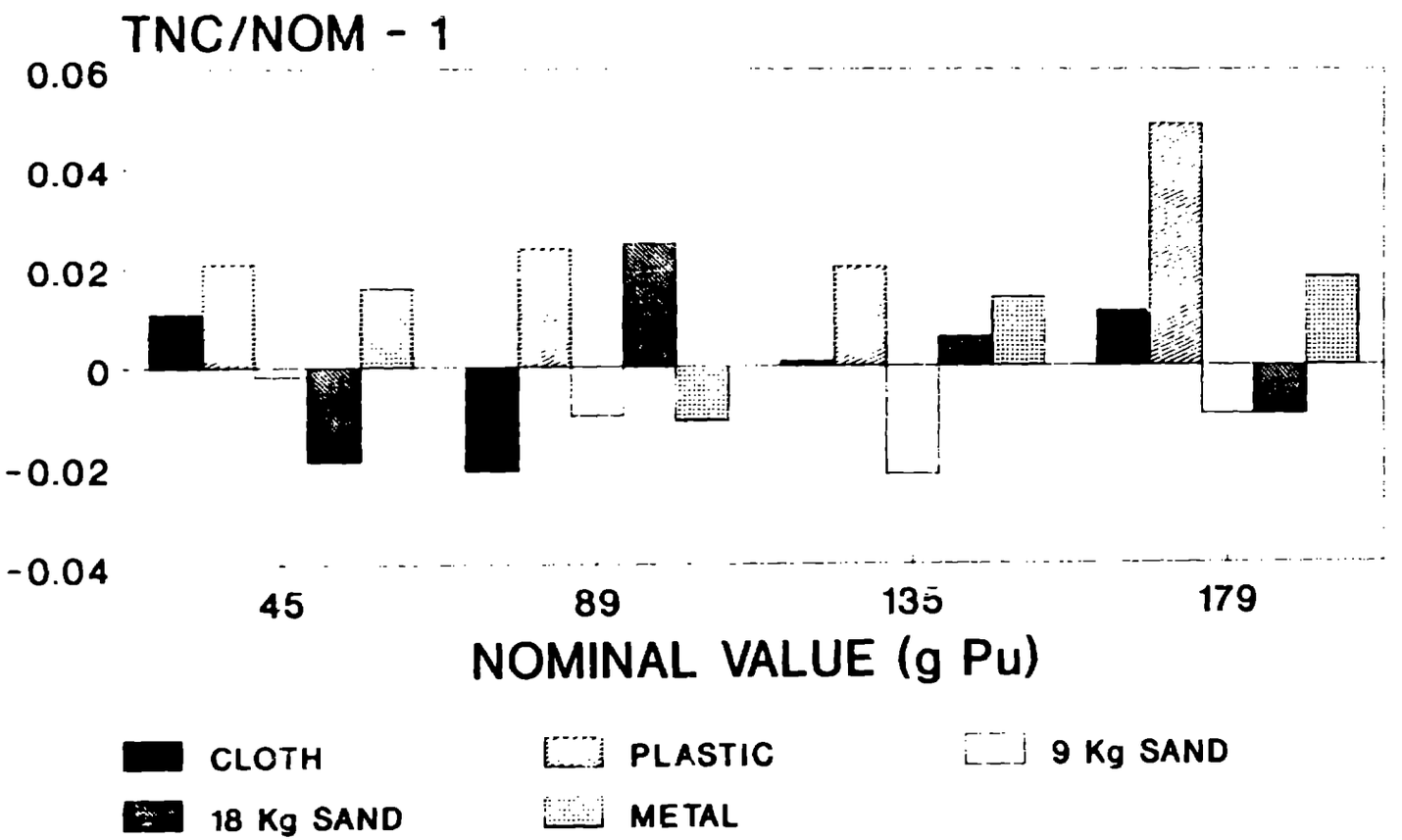

Figure 2

ACRONYMS

LANL - Los Alamos National Laboratory

MSE - Molten Salt Extraction

DOR - Direct Oxide Reduction

TNC - Thermal Neutron Counter

SGS - Segmented Gamma Scanner

C/I - Calorimetry plus isotopics analysis

IDMS - Isotopic Dilution Mass Spectroscopy

NDA - Non-Destructive Assay

TRU - Transuranic

NON - DESTRUCTIVE MEASUREMENT OF SOLID PLUTONIUM WASTE AT LOS ALAMOS NATIONAL LABORATORY (LANL)

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TRU WASTE AT LANL

- TRU WASTE IS GENERATED FROM METAL FABRICATION AND PYROCHEMICAL & AQUEOUS PROCESSING OF PU SCRAP
- PU MATERIALS WHICH REQUIRE NDA ANALYSIS INCLUDE:
  - Pu Oxide from Burned Residues
  - Pu Salts from Production/Reduction
  - Pu Salts from Metal Purification
  - Metal Reduction Slags
  - Ceramics
  - Auxiliary Equipment

- NDA OF TRU WASTE IS COMPLICATED BY
  - Variety of Chemical Forms, Matrix Compositions, Physical Sizes, and Isotopic Proportions
  - Generally Heterogeneous
  - Representative Standards Frequently Do Not Exist

NDA WASTE ASSAYS AT LANL

- Segregation According to Chemical Form, Isotopic Composition, and Matrix Content
- Low Density Wastes - SGS
- Medium and High Density Wastes - TNC
- Problem Materials - Studies

NDA MEASUREMENT STUDIES

- Hydrofluorination Wastes
- Molten Salt Extraction Wastes
- Direct Oxide Reduction Salts
- Moderation Effects in TNC Drum Counters

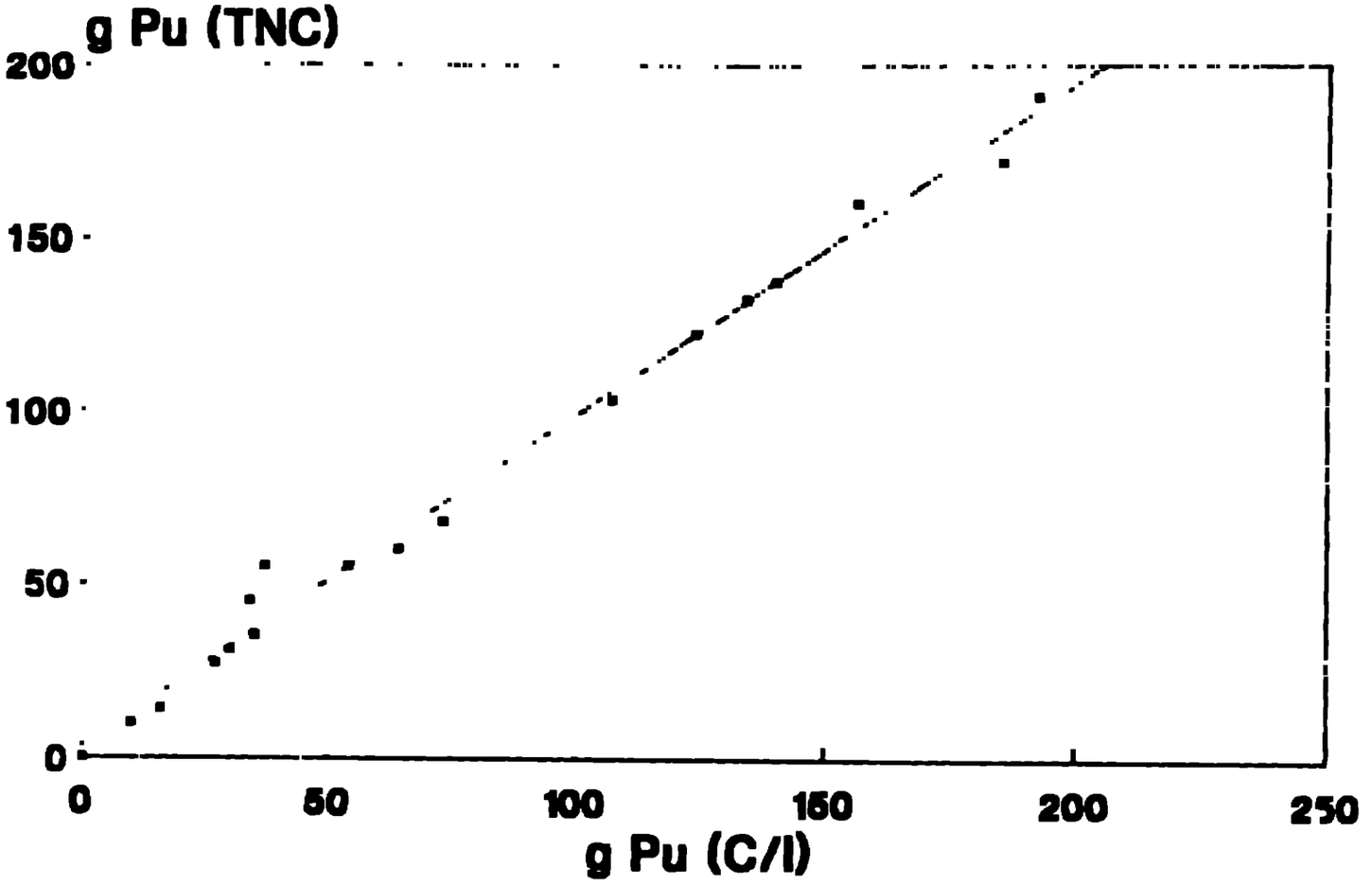
STUDY: HYDROFLUORINATION WASTES

- Slag and Crucible Remnants Resulting from PuF_4 Reduction
  - Contains CaF_2, PuF_4, MgO, Pu shot
- Comparative SGS, TNC, C/I Study to Determine Bias

STUDY: HYDROFLUORINATION WASTES

- Results
  - SGS Measurements Biased Low by Unpredictable Amounts
  - TNC Measurements Biased High, but Bias is Uniform from 0 g to 200 g of Pu
- Simple, Linear Bias Correction Factor Can Be Used With TNC Measurements

HYDROFLUORINATION WASTES
Corrected TNC Data



STUDY: MOLTEN SALT EXTRACTION WASTES (MSE)

- Process Reduces Am Content in Pu Metal
  - Salt Residue Contains KCl, NaCl, MgCl, $PuCl_3$, $AmCl_3$, MgO Crucible Shards, Pu Shot
- Comparative IDMS, SGS, TNC, C/I Measurements Performed to Study Bias

STUDY: MOLTEN SALT EXTRACTION WASTES (MSE)

ITEM	IDMS (g Pu)	Conventional SGS / IDMS	Lump-Corr SGS / IDMS	TNC / IDMS	C/I / IDMS
1	55.56	1.01	-	-	1.03
2	90.39	1.00	0.95	1.35	1.03
3	141.21	0.91	-	-	0.99
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8	372.73	0.98	0.95	2.12	1.10
9	408.58	0.92	0.97	2.20	1.06
	AVG	0.96	0.96	1.71	1.04

STUDY: MSE SALTS

- Results
  - All 4 NDA Techniques Are Biased
  - Additional C/I Studies Underway
  - Current Practice
    SGS: < 50 g
    C/I: > 50 g

STUDY: DIRECT OXIDE REDUCTION RESIDUES (DOR)

- Pu Metal Production Step
  - Residue Contains CaO, CaF2, CaCl2, low Am 241
  - Too Dense for SGS Analysis
  - Presence of F makes TNC Measurements Questionable
- Comparative IDMS, TNC Study Performed to Quantify Bias

STUDY: DOR RESIDUES

- Results
  - TNC Measurements Biased High by an Average of 3.5 %
  - TNC Measurement Uncertainty (Random Error) is 5 % to 10 %
  - Additional TNC Studies Underway

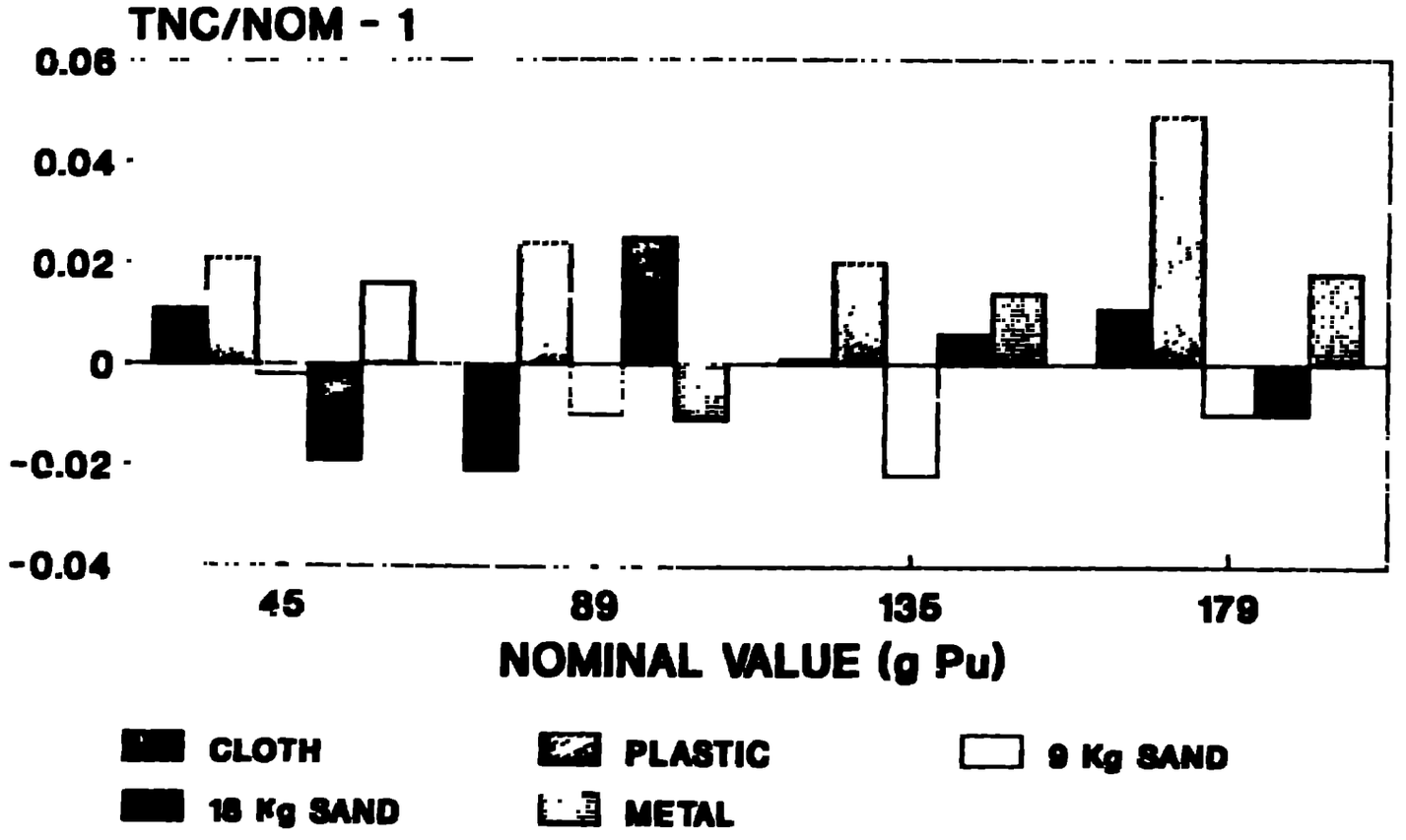
STUDY: MODERATION EFFECTS IN TNC DRUM COUNTERS

- Waste Packaged in Drums Generally Has Substantial Quantities of Matrix Material
- Neutron Moderation from Some Matrices Can Affect TNC Assays
- Study Performed to Quantify These Effects

STUDY: MODERATION EFFECTS IN TNC DRUM COUNTERS

- In the Study, Materials Representing Major Solid Waste Categories Generated at LANL Were Simulated
  - High Density Matrix: 35 Kg Steel
  - Medium Density Matrix: 9 Kg & 18 Kg Sand
  - Low Density Matrix: 8 Kg Plastic
    2 Kg Cloth
- Pu Standards from 45 g to 179 g Were Loaded into Drums, Surrounded by Matrix Material, and Measured

MODERATION EFFECTS IN TNC DRUM COUNTER



STUDY: MODERATION EFFECTS IN TNC DRUM COUNTERS

- Results
  - No Bias for Cloth, Sand, or Metal
  - Small, High Bias for Plastic (< 3 %)
- Outcome
  - Bias is Quantified
  - Present Drum TNC Usage Justified

UNRESOLVED WASTE MEASUREMENT ISSUES AT LANL

- Ongoing Problems That Affect Waste Measurements Here
  - Changes in Instrument Response: RF Noise, Variations in Electrical Power, Deterioration of Detector Response Over Time, Seasonal Variations in Calorimeter Baseline Voltages

UNRESOLVED ISSUES - CONTINUED

- Modifications in TRU Processing
  - Has Led to High Concentrations of Pu Shot, Altered TRU/Salt Matrix Ratios, Increased Bulk Densities in TRU Waste - All Affected NDA Measurements
  - Modifications Were Done Without Warning Waste Measurement Personnel

UNRESOLVED ISSUES - CONTINUED

- Packaging Effects: Variations in Size, Shape, Thickness, and Composition of Containers Can Lead to Measurable Effects in Waste Assays
  - Container Attenuation in SGS
  - Moderation Effects in TNC
- Matrix Effects: Am 241 Content, Pu Shot Quantity, Pu/Salt Matrix Concentration May Vary Depending on Completeness of Chemical Reaction

UNRESOLVED ISSUES - CONTINUED

- Changes in Calibration Standards
  - Homogeneous Standards: Will Stratify If Not Shaken Regularly
  - Heterogeneous Standards: Pulverization of Lumps Due to Handling May Lead to Redistribution of Pu

SOLUTIONS

- New Technologies May Help But Are Unproven
  - Lump Corrected SGS
  - Improved Neutron Self-Interrogation Techniques
  - TNC Moisture Content Corrections
  - Flattened Response (Energy and Spatial) Neutron Counters
  - New Counters That "Measure" (Alpha, n) and Multiplication Effects
- Monitoring and Evaluation of Measurements