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TITLE A COMPARISON OF CONVENTIONAL AND PROTOTYPE NONDESTRUCTIVE MEASUREMENTS ON MOLTEN SALT EXTRACTION RESIDUES

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A COMPARISON OF CONVENTIONAL AND PROTOTYPE NONDESTRUCTIVE MEASUREMENTS ON MOLTEN SALT EXTRACTION RESIDUES

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ABSTRACT

Fourteen molten salt extraction residues were assayed by conventional and prototype nondestructive assay (NDA) techniques to be compared with destructive chemical analysis in an effort to identify acceptable NDA measurement methods for this matrix. NDA results on seven samples and destructive results on four samples are presented.

INTRODUCTION

Impure plutonium metal is routinely processed by molten salt extraction (MSE) to reduce the amount of americium in metal product. A by-product of this operation is a salt residue composed of KCl, NaCl, MgCl, $PuCl_3$, and $AmCl_3$ often containing shards of MgO crucible. Historically, these residues have been measured using nondestructive techniques with little confidence. The large quantities of nonmetallic ^{241}Am in these residues frequently generate high neutron count rates due to (alpha,n) reactions. The salts in this study ranged in Am content from a few thousand ppm to several grams. The large count rates cannot be handled by the preamplifiers in conventional thermal neutron coincidence counters and make discrimination between Pu coincidence neutrons and Am (alpha,n) neutrons difficult. Furthermore, assays made with conventional segmented gamma scan and gamma isotopic instruments were predicted to be biased due to the heterogeneous nature of these salts and the wide variability in the Pu to bulk ratios.

The plutonium in MSE residues can be recovered either aqueously or pyrochemically. Recovery of plutonium using both of these methods has increased recently. Due to this

increased, a better understanding of the capabilities of conventional nondestructive techniques to measure plutonium in this matrix is needed. Hence, an evaluation effort was planned to determine the accuracy of conventional NDA methods in the assay of MSE salts. This effort has also provided an opportunity to observe the performance of prototype NDA equipment that has recently become available at the Los Alamos Plutonium Facility.

Fifteen individuals from four technical groups at the Los Alamos National Laboratory and individuals from Rockwell International Rocky Flats Plant have participated in the study. This effort has taken almost one year to perform and some of the destructive assays have yet to be completed. One focus of this study is to improve in-house accountability on MSE residues and to identify assay methods that would be acceptable for determining receiver's values for this matrix from off-site sources using standard NDA methods at Los Alamos and Rocky Flats. Another focus is to test and evaluate the performance of prototype NDA instruments and evaluate the performance of different gamma spectrum interpretation software packages on this historically difficult to measure residue. Specifically, these prototype system studies apply to recent improvements which have been made in a segmented gamma scan system, a thermal neutron coincidence counter, and in the software of a gamma isotopic system that supports the calorimeters at the Los Alamos Plutonium Facility. An unexpected spin-off of this study has been the investigation into difficulties in calorimetry assay of complex materials like MSE residues. Los Alamos has a large R&D capability for designing, building, and testing prototype NDA equipment. This capability exists in proximity to the Los Alamos Plutonium Facility and the analytical chemistry group. These combined elements provide a unique opportunity to perform studies of the nondestructive and destructive assay of difficult to measure materials like pyrochemical residues.

NDA TECHNIQUES STUDIED

Fourteen containers of molten salt extraction residues were selected for study. Seven of these salts originated at the Los Alamos Plutonium Facility and seven originated at the Rockwell International Rocky Flats Plant. At this time, we report data on the seven Los Alamos salts. A full report on all fourteen salts will be available in 1988.

Measurements With Conventional NDA Systems

* Multiple measurements with the conventional segmented gamma scan system
* Multiple assays with gamma isotopic measurements using MUDPI[1] software coupled with calorimetry.
* Multiple measurements with the large water shielded thermal neutron counter. (We do not report these values because this system uses a preamplifier that is too slow to handle the high neutron count rates. Consequently two of the items could be assayed.)

Measurements With Prototype NDA Systems

* Multiple measurements with an advanced segmented gamma scan system.[2]
* Multiple measurements with an upgraded thermal neutron coincidence counter using the self-interrogation approach.[3]
* Multiple measurements with gamma isotopic measurements using GRPAUT[4] software coupled with calorimetry.

DISSOLUTION AND SOLUTION QUANTIFICATION

To obtain reference values, the materials were analyzed by destructive techniques. Bulk dissolutions of four salt items were performed by personnel in the aqueous processing area of the Los Alamos Plutonium Facility. Entire salt items were dissolved in 1M HCl with solution volumes ranging from 7 to 10 liters. Three of the four salt items contained crucible pieces that would not dissolve and all four of the items had some insoluble solids. Residual crucible pieces and undissolved solids from each dissolution were assayed separately by thermal neutron coincidence counting and ranged in value from 0.5 to 1.2g Pu. The Am concentration in these residues was sufficiently low that no count rate effects were observed.

Samples of solution were submitted for plutonium analysis by isotopic dilution mass spectroscopy (IDMS) by the Los Alamos analytical chemistry group.

Due to the large solution volumes and the fact that all of the crucible did not dissolve, we estimate the overall combined uncertainties in the summed IDMS and residue Pu values reported in Table I to be +/-3%. Alternative destructive methods are currently being explored for the remaining 10 residues to reduce the overall uncertainty in the reference Pu values.

PROBLEMS ENCOUNTERED

As outlined above, calorimetric measurements were made on these items. More than 10% variation was observed in some of the power (watts) values obtained from repeated measurements on a single item. These large variations were quite unexpected.

Experiments were designed by Sedlacek et. al.[5] to evaluate this phenomenon. It was determined that predictive watts values vary by up to 10%. Even watts values indicated by the software as equilibrium values can vary by +/-10% if a true equilibrium has not been reached. This variance is likely due to the poor heat conductivity of the salt and crucible matrix. We found it necessary to leave the sample in the calorimeter for over 24 hours to measure a true heat content. For shorter measurement times the calorimeter had not reached equilibrium, but the computer program running the calorimeter "thought" that equilibrium had been achieved because of the slow rate of temperature change. Only multiday equilibrium watts values are used in calculating plutonium values that are compared to the other nondestructive and destructive values reported here. These long assay times introduced an unexpected delay in the overall study.

RESULTS

The results of the various assays are shown in Table I. Only four of the destructive assays are complete at this time. The prototype thermal neutron counter Pu values based on ^{240}Pu as determined by both GRPAUT and IDMS are shown.

TABLE I.

Measurement Comparisons

Sample	Prototype* TNC GRPAUT ISO		Prototype TNC IDMS ISO		Advance SGS		Conventional Calorimetry SGS + GRPAUT		Calorimetry + MUDPI		IDMS* Pu
	g Pu	+/-%	g Pu	+/-%	g Pu	+/-%	g Pu	+/-%	g Pu	+/-%	g Pu	+/-%	
XBLP278	79	2			78.6	0.8	85.2	1.3	92.9	1.6	93.2	9.9	
XBLP120	94	2	93	2	97.1	0.5	97.2	0.3	109.6	0.3	106.0	3.4	111.6
XBLP270	102	2	98	2	96.4	0.6	99.0	2.0	115.3	1.8	100.5	0.6	99.0
XBLP267	117	2	110	2	126.7	0.9	122.5	3.1	144.1	1.2	141.5	12.5	125.5
XBLP121	140	2			141.0	0.3	131.9	2.6	169.1	2.8	155.7	12.9	
XBLPS300	197	2	199	2	177.3	0.7	178.2	2.3	197.4	1.0	199.0	0.5	198.8
XBLPS301	271	2			221.1	6.3	206.3	2.4	249.4	0.7	242.4	3.1	

* Includes TNC assay of residue

DISCUSSION

In general, the calorimetry plus nondestructive gamma isotopic techniques tend to give a higher plutonium value than either the prototype neutron counting or gamma techniques. The magnitude of the variations between values for a given sample range from +/- 5.8% to +/-13.6%.

For four of the seven samples the combined destructively determined chemical assay and TNC Pu values are given in Table I. The other destructive analyses have yet to be completed.

Due to the limited number of destructive analyses completed at this time, no evaluation of the superiority of one method over another is yet apparent.

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