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EVALUATION OF THE NEUTRON SELF-INTERROGATION APPROACH FOR ASSAY OF PLUTONIUM IN HIGH-α,n MATERIALS

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ABSTRACT

Neutron self-interrogation is a proposed method for assay of plutonium in bulk materials with very high α,n activity. The simple assay approach assumes that neutron multiplication for the calibration standards is the same as that for the bulk items. Efforts to use bulk properties to determine corrections to the calibration for changing multiplication have been initiated. Self-interrogation assays of bulk pyrochemical residues have been performed. Comparison with tag values obtained by difference gives poor agreement. Comparison with tag values obtained by dissolution and destructive analysis gives agreement at the 10% (1σ) level with no corrections for changing package dimensions or matrix amounts. The agreement improves by a factor of 2 or more if a bulk correction factor (derived from a packaging matrix study with standards) is applied.

I. INTRODUCTION

Coincidence counting of correlated neutrons that arise from the spontaneous fission of the fertile isotopes of plutonium is an approach to the bulk assay of plutonium of known chemical form. As the plutonium mass increases, neutron multiplication raises the coincidence signal above the linear response for a given effective fertile (^{240}Pu eff.) mass. For pure metallic plutonium all neutrons originate from fission and are mostly correlated.

The measured rates of total neutron coincidences (reals (R)) and total neutrons (totals (T)) can be used[1] to solve both for multiplication (and hence for a correction to R for the induced fission component R(IF)) and for the spontaneous fission reals component R(SF), which is directly proportional to the ^{240}Pu eff mass. However, for impure metal or for compounds of plutonium, α,n reactions add an additional component T(α) to the totals rate. Although uncorrelated, the α,n neutrons can be the source of an additional induced-fission component of the reals rate. When the material is relatively well characterized (for example, when the plutonium is in pure oxide form with a known americium content), the ratio of the α,n to spontaneous-fission components of the totals rate [T(α)/T(SF) or α] can be calculated and used, along with the measured R and T, to correct for multiplication and to solve for ^{240}Pu-eff. This approach[2] is applied internationally and domestically in the verification of bulk quantities of nuclear materials.

Most forms of plutonium in scrap recovery operations, and many forms of plutonium elsewhere, are impure and poorly characterized chemically. Residues from the pyrochemical recovery processes are among the most outstanding examples. Furthermore, certain residue and product categories, such as the spent salt from americium molten salt extraction (MSE), contain large amounts of americium (typically in the few-percent range for low-burnup materials) and α values for these materials are typically 15 or greater. The resulting high totals neutron rates cause R(IF) to approach and often exceed R(SF).

For such materials, multiplication corrections based on R and T measurements cannot be applied because of the large, unknown, and variable α,n component in T. The use of other conventional destructive and nondestructive assay methods is complicated by physical and chemical inhomogeneities, high americium content, and high contact radiation dose rates characteristic of these residues. The pyrochemical residues are often shipped between facilities or stored for extended periods of time. [illegible] determined by difference when a [illegible] measured result would be desirable for [illegible] control purposes.

The methodology of neutron self-interrogation[3] (SI) is a suggested approach to the [illegible] of bulk plutonium-bearing materials with high α,n rates.[4] Because of the extremely high neutron rates, the neutron coincidence counting of high-α,n plutonium bulk residues has not been possible until recently with the introduction of fast-counting upgrades[5] to the traditional analog circuitry used with ^{3}He proportional counters. The high-level neutron coincidence counter, the HLNC-II,[6] is a commercially available well counter that is appropriately upgraded. The equivalent upgrades have also been installed on several of the in-line thermal neutron counters (TNCs) at the Los Alamos Plutonium Facility, TA-55. One of these TNCs and an off-line HLNC-II are currently employed at TA-55 in an evaluation of the neutron self-interrogation approach for the assay of plutonium in high-α,n materials.

This paper documents the progress of the evaluation of the SI method for bulk plutonium assay. The work is the joint effort of the Los Alamos Safeguards Assay Group and the Plutonium Metal Technology Group and relies heavily on the nuclear materials processing, characterisation, and handling capabilities of the research and development organisation at TA-55.

II. SI ASSAY METHODOLOGY

For bulk plutonium-bearing material of relatively constant fissile density and dimensions, the magnitude of R(IF) is a function of both the fissile content and the intensity of the neutron source. The totals rate is a measure of the neutron source intensity. Therefore, the ratio R(IF)/T is a function of the fissile content. The dependence of this function on α decreases as the ratio of uncorrelated (α,n) to correlated (fission) neutrons increases. Therefore, the fissile content of materials such as the spent MSE salts (for which the α,n neutron rate is typically more than 90% of the totals rate) can be treated as a simple function of R(IF)/T, independent of the α,n source term.

The simple SI assay requires a knowledge of two functions. One of these describes the dependence of the fissile mass on R(IF)/T. The other describes the dependence of R(SF) on the fertile mass. The inverse of the second function is the calibration function for the effective fertile assay based on the measured (multiplication corrected) spontaneous fission reals rate. Both functions can be determined with standards. The iterative use of these functions in the simple SI assay method can be described graphically using Fig. 1. The measured R/T ratio is used to obtain a first approximation to the effective fissile mass, g ^{239}Pu-eff, from the graph at the right side of Fig. 1. This is used to solve for the plutonium mass, g Pu, with the formula[7]

TOTALS $\quad T = T(SF) + T(\alpha,n) + T(IF)$

REALS $\quad R = R(SF) + R(IF)$

$R(IF) = R - R(SF)$

MULTIPLICATION $\quad \frac{R(IF)}{T} \sim {}^{239}\text{Pu-eff}$

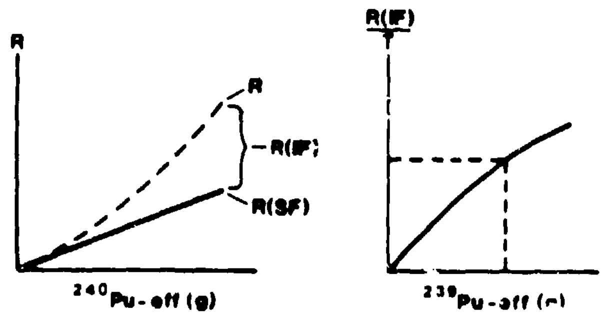

Fig. 1. Graphic illustration of the simple SI assay methodology. The plot at the right is the (measured) ratio R(IF)/T vs the effective fissile plutonium mass. The plot at the left is the (inferred) R(SF) vs the effective fertile plutonium mass. The plots are used iteratively to convergence.

$$g\ {}^{239}\text{Pu-eff} = g\ \text{Pu} \cdot (0.738 \cdot f_{238} + f_{239} + 0.502 \cdot f_{240} + 1.367 \cdot f_{241} + 0.407 \cdot f_{242} + 0.539 \cdot f_{241,Am}) \quad . \qquad (1)$$

where the f quantities are the known isotope weight fractions. (For low-burnup plutonium, only an approximate knowledge of these isotopic fractions is required because of the dominance of ^{239}Pu, although the effect of variable and unknown amounts of americium at the few percent level is clear.) The plutonium mass is used to solve for the effective fertile mass.[1]

$$g\ {}^{240}\text{Pu eff} = g\ \text{Pu} \cdot (2.54 \cdot f_{238} + f_{240} + 1.69 \cdot f_{242}) \quad . \qquad (2)$$

This is used to obtain R(SF) from the graph at the left side of Fig. 1. The R(SF) result is used to update R(IF) as follows:

$$R(IF) = R - R(SF) \quad .$$

Then R(IF) is divided by T to obtain the next approximation to R(IF)/T and hence g ^{239}Pu-eff. Convergence at the 1% level occurs after about five iterations. Note that Eq. (3) is valid when α is very large.

III. PACKAGING AND MATRIX CONSIDERATIONS

The simple formalism described above can be applied when the α,n rates are very high and when the package dimensions remain fixed. However, if the shape of the bulk package changes, or if matrix material is removed (or added) to concentrate (or dilute) the plutonium, the multiplication changes, and R(IF)/T will change for a given fissile mass.

The MSE salts, as well as other pyrochemical residues generated at Los Alamos, are stored in four different can sizes. Furthermore, the bulk masses typically vary from 1 to 3 kg, although the plutonium mass rarely exceeds 200 g. Because of high radiation dose rates, the spent salts are broken out from the crucibles with minimal handling. The containers of spent salt held chunks ranging in size from small grains to large (~10-cm) irregularly shaped pieces. Also packed with the salt (more often than not) are the broken pieces of magnesium oxide crucible. Finally, residues shipped to Los Alamos from other sites arrive in their own characteristic containers and display other packaging and matrix dissimilarities relative to the Los Alamos-generated residues.

IV. SIMPLE MODEL OF PACKAGING AND MATRIX EFFECTS

The calibration for the SI assay consists of two functions described graphically in Fig. 1. The quantity R(IF)/T for a given ^{239}Pu-eff mass will differ from the result determined with the standards if the bulk material is in a different-size package from that of the standards and/or if the volume of matrix in the bulk material differs from that of the standards. It is desirable to be able to correct the calibration based on the observed bulk properties so that a single calibration (obtained from one set of standards), appropriately corrected, can be used for all package dimensions and matrix quantities.

A macroscopic expression is derived for this purpose. For bulk fissile material, the fraction of neutrons produced within the bulk mass that induce fission in a single collision is

$$f_I = 1 - e^{\sigma_{n,f} \bullet \rho \bullet \chi}$$

$$\approx \sigma_{n,f} \bullet \rho \bullet \chi \quad , \qquad (4)$$

where $\sigma_{n,f}$ is the microscopic cross section for induced fission, ρ is the fissile atom density, and χ is the neutron path length. The average chord length through an object of arbitrary shape[8] is

$$\bar{\ell} = 4 \bullet v/a \quad , \qquad (5)$$

where v and a are the volume and surface area, respectively, of the object. For a cylindrically shaped object, $\bar{\ell}$ can be expressed as

$$\bar{\ell} = 2 \bullet h \bullet r/(r + h) \quad , \qquad (6)$$

where r and h are the radius and height, respectively, of the cylinder. The fissile atom density for a cylindrical package is

$$\rho = M_f \bullet N_o/A \bullet \pi \bullet r^2 \bullet h \quad , \qquad (7)$$

where M_f is the fissile mass, and N_o and A are Avogadro's number and the atomic mass (239 for low-burnup plutonium), respectively. Assume that χ is proportional to $\bar{\ell}$. Substituting Eqs. (6) and (7) into Eq. (4) (where $\bar{\ell}$ replaces χ) gives the fraction of neutrons produced within a cylindrical bulk mass that induce fission in a single collision:

$$f_I \propto 2 \bullet \sigma_{n,f} \bullet M_f \bullet N_o/A \bullet \pi \bullet r(r + h)$$

$$= (2 \bullet \sigma_{n,f} \bullet M_f \bullet N_o/A \bullet \pi) \bullet g_I \quad , \qquad (8)$$

where

$$g_I = [r(r + h)]^{1} \qquad (8a)$$

is the geometry factor.

For two cylindrical bulk items, 1 and 2, with the same M_f but different r and h (because of different containers and/or matrix quantities), the ratio of the induced-fission probabilities is

$$f_{I,1}/f_{I,2} = g_{I,1}/g_{I,2}$$

$$= CF \quad , \qquad (9)$$

The quantity CF is a correction factor based on bulk geometries that is applied to calibration function 1 (obtained with standards in container 1 that were mixed with matrix to fill height 1) to give calibration function 2 (corresponding to a different container diameter and/or matrix amount).

V. STANDARDS MEASUREMENTS

Two sets of standards were prepared for use in the calibration and evaluation of the SI assay. Because the special nuclear material (SNM) used for these standards was a recent product of scrap recovery, the americium levels were low. The high α,n rate requirement was achieved with PuF_4, and in some cases, the standards contained PuO_2 admixtures. The major components of the standards were KCl and NaCl salts in equal molar amounts, typical of many pyrochemical salt residues including most MSE salts generated at Los Alamos. All SNM used for the standards as well as all process materials reported herein consisted of low-burnup (6% ^{240}Pu) plutonium. Therefore the calibration data and subsequent assay results are presented here vs total plutonium mass, which is directly proportional to total fissile mass with a negligible error (<0.5%, 1σ).

The "original" standards,[9] 1-8, consisted of weighed amounts of PuF_4 and, in four cases, PuF_4 and PuO_2 (to vary the magnitude of the α,n contribution to the totals rate) mixed with 300 g $MgCl_2$ (the MSE oxidizing agent) and enough KCl and NaCl to achieve a total mass of 1000 g. Standards 1-4 contained no PuO_2. The percent plutonium contributed by the PuO_2 to the total plutonium mass in standards 5-8 was 18, 24, 62, and 51, respectively. The salt matrix mass is low compared with typical bulk salt masses observed for spent MSE salts (>2000 g). The original standards were packaged in No. 10 slip-lid stainless steel cans (pictured in Fig. 2). The average ratio of the stoichiometric plutonium masses of the original standards to segmented gamma scan assays (precise to 2.5%, 1σ) performed on these standards was 1.03 ± 0.01 (1σ).

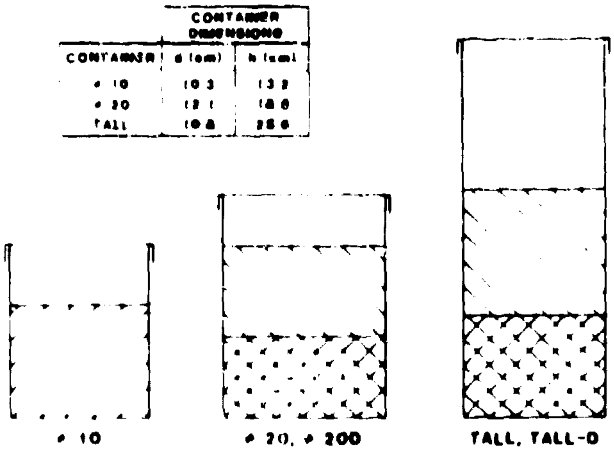

Fig. 2. Drawings of the containers used in the standards study of packaging and matrix effects. The container dimensions are given. The approximate fill heights for the standards, before and after dilution (D), are shown.

The original standards were neutron counted in the HLNC-II.

Because the original standards had been shipped to another site temporarily, after completion of measurements with the HLNC-II (and before the TNC upgrade), a second set of standards was prepared to provide additional calibration data for the HLNC-II and for the upgraded TNC and also for use in a study of packaging and matrix effects. The "new" standards, 9-14, contained only PuF_4, KCl, and NaCl, and were packaged initially in the same No. 10 cans used for the original standards. The masses of the salt matrix were chosen to achieve a total mass of 1000 g initially. The PuF_4 used for the new standards was recently prepared by hydrofluorination of PuO_2. The batch was mixed and three samples were taken for destructive analysis before the PuF_4 was weighed out for the standards. The sampling and weighing were carried out on the same day in a dry air atmosphere. Table I gives the plutonium masses for the standards computed from stoichiometric weight fractions applied to the weighed quantities of PuF_4 as well as those obtained using the weight fractions from destructive analysis. The agreement is excellent. (Refer to Table I.) For additional verification of the reference values, calorimetry and gamma isotopics[10] measurements were performed on the individual bulk standards. Agreement with the reference values to ~0.5% (the approximate uncertainty in the nondestructive assay result) is observed in all but one comparison, as shown in Table I. Also, the nondestructive assay result for standard 9 (for which there is no destructive analysis reference value) confirms the original result obtained by weighing and stoichiometry. The new standards were measured in both the HLNC-II and the TNC, but only the HLNC results are reported here.

The original and the new standards were neutron counted in the No. 10 containers. The new standards were transferred first to No. 20 and then to "tall" containers (Refer to Fig. 2.) and recounted. Finally, the new standards were diluted with an additional 1000 g of NaCl and counted in the tall and No. 20 cans again.

Using the reference values for plutonium mass and the plutonium isotopics results from the gamma spectrometry measurements, the R(SF) values are computed for each standard using Eq. (2) and

$$R(SF) = 18.14 \bullet g\ ^{240}Pu\text{-eff} \quad . \qquad (10)$$

where the constant, 18.14, is based on the knowledge[9] of the HLNC II coincidence count rate for ^{240}Pu spontaneous fission. The R(IF) is obtained from Eq. (3), and R(IF)/T is plotted vs grams of plutonium for all of the HLNC II standards measurements in Fig. 1.

TABLE I

PuF_4 STANDARDS

Std	Grams Plutonium (1σ) Weight[a]	Reference[b]	CAL-ISO[c][d]	Weight / Reference (1σ)	CAL-ISO / Reference (1σ)
1	25				
2	75				
3	125				
4	200				
5	50				
6	75				
7	100				
8	150				
9[e]	25.0 (0.2)		25.2 (0.1)		
10	49.6 (0.2)	49.6	49.7 (0.2)	1.000 (0.004)	1.002 (0.004)
11	99.3 (0.2)	99.2	102.0 (0.4)	1.001 (0.002)	1.028 (0.004)
12	149.0 (0.2)	149.0	149.0 (0.8)	1.000 (0.002)	1.005 (0.004)
13	198.6 (0.2)	198.6	199.8 (0.7)	1.000 (0.001)	1.006 (0.004)
14	297.8 (0.2)	297.9	297.2 (1.1)	1.000 (0.001)	0.998 (0.004)
			Average	1.000	1.008
			Standard Deviation	0.001	0.012

[a]Stoichiometry (for pure PuF_4) applied to the weighed amounts of PuF_4 to give grams of plutonium. The stoichiometric result is 0.7548 g Pu per gram PuF_4.

[b]Reference values obtained from controlled potential coulometry performed on three samples taken from the PuF_4 batch used to prepare the standards. The coulometry result of 0.7507 g Pu/g PuF_4 (1σ = 0.0008 g Pu/g PuF_4) is applied to the PuF_4 weights to give grams of plutonium.

[c]Equilibrium calorimetry measurements of the individual bulk standards.

[d]Plutonium gamma-ray isotopics (using MUDPI[13]) measurements of the individual bulk standards.

The combined results c and d give grams of plutonium.

[e]Standard 9 is actually standard 1, which was included in the subsequent study with the new standards (10-14).

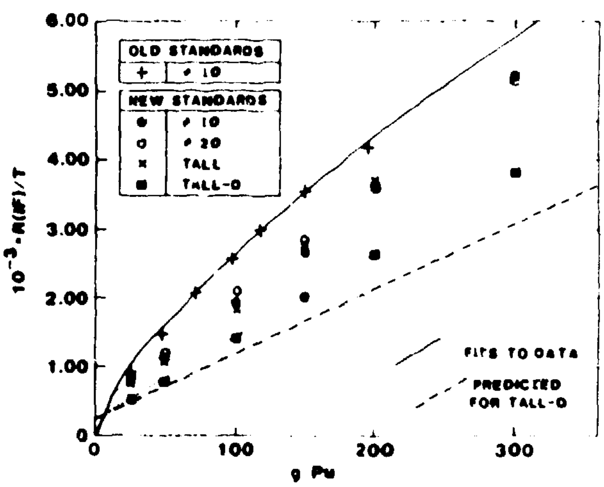

Fig. 3. The R(IF)/T results vs plutonium mass in the PuF_4 standards. The fit to the old standards data (heavy solid line) is the calibration function used to assay the process materials. The fit to the new undiluted standards data (thin solid line) is corrected with the simple model for the dilution effect to give the dashed line. The fit to the new diluted standards data is the dotted line.

The radius and fill height (r and h, respectively) of each new standard in each of the five package/matrix configurations are given in Table II. These dimensions are used to compute CF [Eq. (9)] with the g_I values defined by Eq. (8a). The CF value in this case is the predicted ratio of R(IF)/T values for a fixed fissile mass in two different packages, one of which is the undiluted standard in the No. 10 can. The predicted ratios are given in Table III. The uncertainties in the predicted ratios are obtained from the uncertainties in the r and h values, as defined in Table II.

Table III also gives the measured CF value, the measured ratio of R(IF)/T for the standard in the No. 10 can to that in each of the other package/matrix configurations. The uncertainty in the measured ratio is obtained from the standard deviation in the mean result of measurements of each package performed on different days.

The predicted and observed values of CF are near unity for the undiluted standards. Although there may be small systematic differences between the observed and calculated results for a given package, they are not large enough relative to the individual measurement uncertainties to help in drawing conclusions about the usefulness of the model. It is encouraging to note that a simple change in container for the same material causes a change (predicted and observed) in the assay signal of ~5%.

The predicted CF values for the diluted standards are large (1.70 and 1.64 for the tall-D and 20-D packages, respectively) and constant within uncertainties for the two D-package types. The observed results are not as large and show significant variation as a function of the plutonium mass. The plotted results in Fig. 3 illustrate the trends. The data points are the values of R(IF)/T vs plutonium mass. The lines through the data are fits. All data for the undiluted new standards were used to obtain a single fit. The R(IF)/T values from this fit were divided by 1.70 (the predicted CF result for the tall-D standards) and this result is plotted as the dashed line. The observed results for the tall-D standards are also plotted with a straight-line fit (dotted) to these points.

The model appears to improve as the plutonium mass decreases. The increasing gap observed (in Fig. 3) between the predicted (dashed) and observed (dotted) CF values as plutonium mass increases is probably largely the result of plutonium self-shielding effects, which are not included in the model. The self-shielding is greatest for the large-plutonium-mass standards in the undiluted matrix. For these standards, the observed dilution effect is partially compensated for by a decrease in self-shielding. Unfortunately, the large variations in matrix amounts achieved by dilution of the standards are realistic for actual pyrochemical residues. To properly quantify the combined effects of package size plus matrix quantity as well as self-shielding and other systematic effects that may result from the changes in matrix quantity and geometry, a microscopic calculation such as a Monte Carlo simulation is recommended.

The data plotted in Fig. 3 show large variations in the observed signal for a given container as the amount of matrix changes. In the absence of a model to correct the calibration for matrix quantity, a calibration is determined from the data for the original standards. This is applied to the neutron counting results obtained with the process materials. The calibration equation (an exponential fit to the + data in Fig. 3) for low-burnup plutonium is

$$g\ Pu = 349.755 \cdot [R(IF)/T]^{1.3711} \quad (11)$$

Equation (11) corresponds to the (inverse of the) right half of Fig. 1 in the graphic description of the SI assay methodology. The left half of Fig. 1 is represented by Eq. (10).

The data for the original standards show curvature at higher masses that could be the result of self shielding for this higher SNM density situation. The SNM density for the original standards is higher than might be expected

TABLE II

DIMENSIONS OF PuF_4 STANDARDS USED IN PACKAGING ST...

	Dimension[a] (cm)									
	No. 10		No. 20		Tall		Tall-D		No. 20-D	
Std	r	h	r	h	r	h	r	h	r	h
9	5.17	9.11	6.07	6.67	5.29	8.26	5.29	18.26	6.07	13.33
10	5.17	9.36	6.07	6.99	5.29	8.89	5.29	18.57	6.07	13.97
11	5.17	8.73	5.07	8.67	5.29	8.41	5.29	18.41	6.07	13.81
12	5.17	8.73	6.07	6.35	5.29	8.10	5.29	18.41	6.07	13.65
13	5.17	8.41	6.07	5.56	5.29	7.78	5.29	16.99	6.07	12.86
14	5.17	7.78	6.07	5.40	5.29	6.83	5.29	15.87	6.07	11.75

[a]The uncertainty (1σ) in the h value, determined from the scatter about a straight-line fit to grams Pu vs h, is 0.25 cm. The uncertainty in r is assumed to be half this amount.

TABLE III

RELATIVE INDUCED FISSION PROBABILITIES

	q_I(No. 10)/q_I Predicted (1σ) Observed (1σ)									
Std	No. 10		No. 20		Tall		Tall-D		No. 20-D	
9	1.00	1.00	1.05 [0.04]	0.98 [0.13]	0.97 [0.04]	1.06 [0.13]	1.69 [0.07]	1.60 [0.18]	1.60 [0.06]	1.41 [0.12]
10	1.00	1.00	1.05 [0.04]	0.96 [0.05]	1.00 [0.04]	1.06 [0.04]	1.68 [0.07]	1.49 [0.07]	1.62 [0.06]	1.39 [0.03]
11	1.00	1.00	1.07 [0.04]	0.91 [0.05]	1.01 [0.04]	1.04 [0.03]	1.74 [0.07]	1.32 [0.03]	1.68 [0.06]	1.11 [0.06]
12	1.00	1.00	1.05 [0.09]	0.93 [0.04]	0.99 [0.04]	0.96 [0.04]	1.74 [0.07]	1.32 [0.02]	1.67 [0.06]	1.20 [0.02]
13	1.00	1.00	1.01 [0.04]	0.90 [0.02]	0.99 [0.04]	0.97 [0.02]	1.68 [0.07]	1.38 [0.02]	1.64 [0.06]	1.20 [0.01]
	1.00	1.00	1.04 [0.04]	1.01 [0.01]	0.96 [0.04]	1.00 [0.02]	1.67 [0.07]	1.16 [0.02]	1.62 [0.06]	1.27 [0.01]
Av	1.00	1.00	1.05	0.96	0.99	1.01	1.70	1.41	1.64	1.25
Std Dev	.00	.00	0.01	0.04	0.02	0.04	0.03	0.11	0.03	0.10

for most process residues, so that a calibration based on these data is expected to underestimate the plutonium mass of the process materials except for those with lower matrix quantities.

VI. MEASUREMENTS OF PROCESS RESIDUES

The neutron SI method is one of several new approaches that offers some promise for assay of plutonium in MSE salts. A study to evaluate these new approaches[11] has provided plutonium reference values for the residues that were obtained by analysis of solution samples from the dissolution of the MSE salts (following the bulk assays performed by each of the candidate methods). Americium analyses were also performed on the dissolved samples. Fourteen MSE salts were involved in the study. To date, four of these have been dissolved to give reference values. However, an additional five MSE salts (not included in the larger study) were dissolved in an effort to test the dissolution and subsequent analysis procedures, and plutonium and americium reference values were obtained for these five. The TNC and the HLNC-II were used to count all of the bulk items before dissolution.

The SI method, calibrated with the data obtained from the original standards counted in the HLNC-II, produced the plutonium assay results given in Table IV. The SI assay results vary with the ^{240}Pu value because a change in ^{240}Pu-eff alters the R(SF) value computed and used in each iteration [in Eqs. (10) and (3), respectively]. The ratios of the SI result to three different tag values [destructive (DA) and nondestructive (NDA) analysis as well as values obtained by difference] are also given in Table IV for each dissolved salt, and these ratios are plotted in Fig. 4 vs plutonium mass. The measured precision of the SI assay for these materials is 2% (1σ). The standard deviation in the ratios (with DA and NDA tag values) is 10%, most of which can be attributed to the SI assay.

TABLE IV

ASSAY RESULTS FOR DISSOLVED MSE SALTS

	SI Assay Result, g Pu (^{240}Pu Isotope Fraction)				TAG (SI[a]/TAG)		
ID	Book Isotopics	GRPAUT[b] Isotopics	DA Isotopics	Av (1σ)	DA[c]	NDA[c]	BY-DIFF
120	93.3 (0.0592)	94.6 (0.0579)	93.3 (0.0593)	93.7 (0.8%)	111.6 (0.84)	106 (0.88)	N/A
267	109.8 (0.0596)	116.9 (0.0512)	110.3 (0.0591)	112.3 (3.5%)	125.5 (0.88)	121 (0.91)	195 (0.57)
270	96.6 (0.0596)	101.[illegible] (0.0550)	98.1 (0.0583)	98.9 (2.8%)	99.0 (0.99)	95 (1.03)	106 (0.93)
300	201.1 (0.0597)	196.6 (0.0614)	198.9 (0.0605)	98.9 (1.1%)	198.7 (1.00)	186 (1.07)	N/A
263	101.9 (0.0555)	N/A	104.1 (0.0535)	103.0 (1.5%)	102.4 (1.02)	109.8 (0.95)	106 (0.98)
264	90.1 (0.0557)	N/A	87.9 (0.0562)	89.0 (1.7%)	108.0 (0.81)	103.6 (0.85)	123 (0.71)
266	311.9 (0.0487)	N/A	289.3 (0.0547)	300.6 (5.1%)	266.0 (1.09)	265.0 (1.09)	140 (2.07)
284	104.1 (0.0622)	N/A	110.3 (0.0577)	107.2 (4.1%)	119.9 (0.92)	121.3 (0.91)	110 (1.00)
286	95.3 (0.0612)	N/A	96.0 (0.0626)	95.7 (0.5%)	89.7 (1.07)	[illegible]	128 (0.75)

[a]The SI assay result obtained with the ^{240}Pu isotopic fraction determined by destructive analysis was used here.
[b]Gamma-ray isotopics designed for plutonium in forms that are chemically inhomogeneous and high in americium.[12]
[c]See Ref. 11 for a description of the DA and NDA methods used on the dissolved samples to give these tag values.

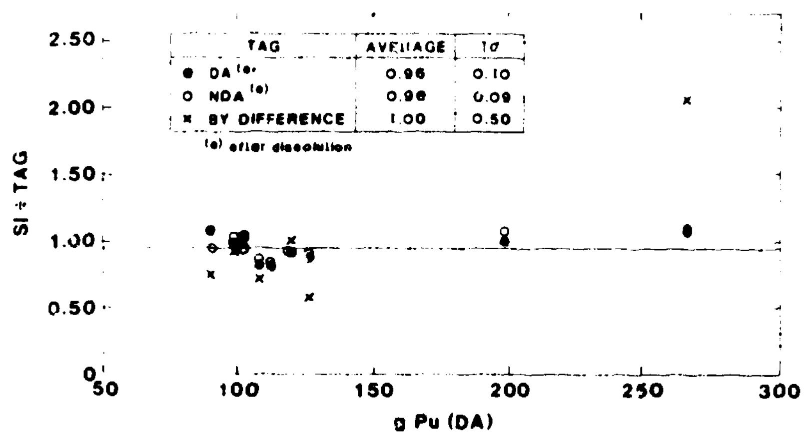

Fig. 4. The ratio of the SI assay result to the tag value vs plutonium mass (determined by destructive analysis of the dissolution samples) for the dissolved bulk MSE salts.

Although 10% is a significant improvement in the 50% result with by-difference tag values, further improvements are desirable for accountability assays.

The dissolution results indicated americium levels of 1% to 3% (relative to plutonium) in the low-burnup MSE salts. This is consistent with expectations based on gamma spectroscopy measurements of bulk salts of this type. Because the 2% ± 1% americium content is a fissile component, the uncertainty introduces an additional (≤0.5%, 1σ) uncertainty in the effective fissile content, g ^{239}Pu-eff [Refer to Eq. (1).], and hence in the SI assay result.

VII. DISCUSSION OF RESULTS

The 10% standard deviation in the SI/TAG ratios (where the tag values are the reference values) for the MSE salts is partly a result of variations in the quantity of salt matrix (and other non-SNM components of the residue such as crucible pieces) in the bulk material within the containers. All bulk MSE salts were assayed in No. 20 cans. Therefore, a correlation between SI/TAG and the container fill height (or perhaps the bulk mass) is expected.

Figure 5 is a plot (large points) of the SI/DA ratios vs bulk amounts (masses and fill heights) of the four MSE salts (those involved in the larger study) for which these quantities were determined before dissolution. If the assay is unbiased, the expected ratio is unity. The ratio shows a decrease vs either mass or fill height, the expected trend for the matrix "dilution" effect examined previously. To determine whether the observed slope is consistent with the magnitude of the dilution effect, the results of the measurements of the new standards (from the matrix and packaging study) are also plotted in Fig. 5 (small points). These are ratios of the SI assay results for the new standards. The denominator in each case is the SI assay for the (undiluted) standard in the No. 10 can because this geometry matches that of the calibration standards and should be immune to the systematic effects of packaging and dilution. The numerator is the SI assay for the repackaged standard (No. 20 and tall cans), undiluted and diluted. The ratios are plotted vs the fill heights of the repackaged standards given in Table II. The results for each package show a systematic trend vs standard mass (defined by the smooth curve that connects the points for each package, where the larger fill heights correspond to lower plutonium masses), but the overall trend vs fill height resembles that for the MSE salts. The average plutonium mass for the four MSE salts is 137 g. The straight line labeled BCF (bulk correction factor) is drawn to intersect the smooth curves Table IV and Fig. 4 between the points for the 100- and 150-g standards. This line is the empirical evaluation of the dilution effect based on the results of the packaging and matrix study with the standards. Correcting the SI assay results with the BCF (as indicated in the lower left of Fig. 5) reduces the standard deviation in SI/DA by a factor of 2 or more. This large improvement provides justification for a continuation of the modeling effort that will generate a calculated correction to the R(IF)/T assay signal based on the observed bulk properties.

Table V gives the neutron counting results for the nine MSE salts for which reference values have been determined. These are used with nuclear data for spontaneous-fission decay (the average neutron multiplicities for ^{240}Pu spontaneous fission and ^{239}Pu induced fission of 2.16

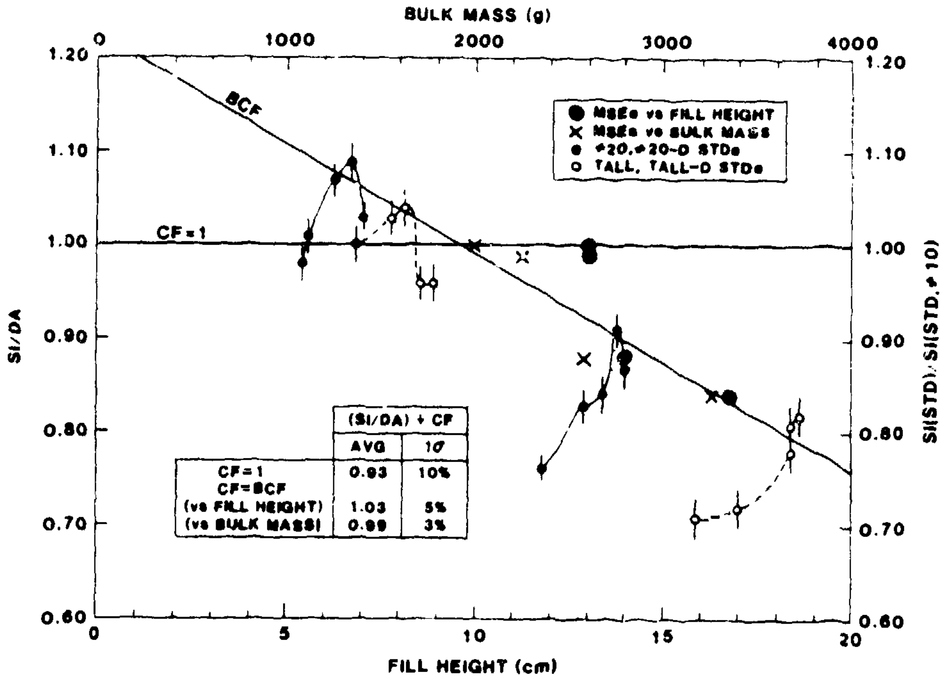

Fig. 5. The large data points plot the ratio of the SI assay result to the DA value vs fill height of the No. 20 container (lower horizontal axis) or vs bulk mass (upper horizontal axis). The small data points plot the ratio of the SI assay result of each new, repackaged standard to the SI assay of the same standard in the No. 10 can vs fill height for the repackaged standard.

and 3.16, respectively, and the ^{240}Pu spontaneous-fission decay rate of 472 $s^{-1}\ g^{-1}$) and with known HLNC-II counting efficiencies [0.175 for totals and 18.14 s^{-1} $(g\ ^{240}Pu\text{-eff})^{-1}$ for reals] to compute T_{RAT}, the ratio of uncorrelated to correlated neutrons. The simple SI method requires this ratio to be large. For most MSE salts, it exceeds 20. For some (especially for ID 300), it is much smaller, and this leads to a significant fraction of fissions that are induced by correlated neutrons. These higher multiplicity induced-fission events cause a positive bias in the simple SI assay. For the ID 300 MSE salt where T_{RAT} is -5 (because this salt is the residue of a second americium extraction of impure metal), the expected bias is -14% relative to the PuF_4 standards (for which T_{RAT} varies between 40 and 80). When T_{RAT} is 20, the expected bias drops to -2%. Because this effect can be quantified once T_{RAT} is known, it is proposed that the simple SI assay be modified to correct the assay signal in an iterative manner in the cases of double-extraction (or other low-americium) spent MSE salts. This effort will be pursued.

VIII. SUMMARY

The simple neutron self-interrogation assay methodology has been applied to spent salts from americium molten salt extraction. For assay results precise to -2%, 1σ, the observed accuracy is -10%, 1σ. The accuracy is shown to improve to 5% or better with corrections for variable amounts of (non-SNM) matrix. Other effects that are shown to contribute to the assay uncertainty are the uncertainty in the ^{240}Pu isotopic content (contributing -2.5%, 1σ), the uncertainty in the amount of (fissile) americium (contributing -0.5%, 1σ), systematic mass effects observed in the results of the matrix/packaging study (contributing -3%, 1σ), and systematic effects resulting from departures from the simple SI assumption of negligible induced fission by correlated neutrons. The latter effect can be addressed with a correction applied to a second iteration of the simple SI assay. The bulk matrix correction can also be incorporated within each SI assay based on the known bulk mass or fill height of the package. Monte Carlo calculations of the packaging/matrix effects are recommended. The accuracy of the corrected assay result should be -5% or better.

TABLE V

COMPONENTS OF R AND T FOR MSE SALTS

CD [g Pu[a]]	T (σ)	R (σ)	R(SF)[b]	R(IF)[c] (σ)	T(SF)[d]	T(IF)[e] (σ)	T(α)[f] (σ)	α[g] (σ)	T_{RAT}[h]
120 [112]	13671 (200)	185 (2)	122	53 (2)	1199	133 (10)	32139 (200)	26.8 (0.2)	21
257 [126]	44130 (200)	230 (1)	137	93 (1)	1149	492 (10)	42489 (200)	31.5 (0.2)	23
270 [99]	28683 (200)	179 (1)	108	71 (1)	1060	375 (6)	27248 (200)	25.7 (0.2)	19
300 [199]	16239 (200)	292 (1)	217	75 (1)	2130	395 (6)	13714 (200)	6.4 (0.1)	5.4
263 (102)	36444 (200)	192 (2)	111	81 (2)	1092	428 (11)	34728 (200)	31.6 (0.2)	23
264 [108]	38783 (200)	179 (2)	118	61 (2)	1156	321 (11)	37306 (200)	32.3 (0.2)	25
266 [266]	35266 (200)	470 (2)	290	180 (2)	2848	949 (11)	31469 (200)	11.0 (0.1)	8.3
284 (120)	23327 (200)	182 (1)	131	51 (1)	1285	269 (5)	21773 (200)	16.9 (0.2)	14
286 [90]	25983 (200)	176 (1)	98	78 (1)	963	412 (5)	24608 (200)	25.6 (0.2)	13

[a] Results from destructive analysis performed on samples of the dissolved salts.
[b] R(SF) = 14.14 * (g ^{240}Pu-eff).
[c] R(IF) = R - R(SF).
[d] T(SF) = 472 * 2.16 * 0.175 * (g ^{240}Pu-eff).
[e] T(IF) = T(SF) * [R(IF)/R(SF)] * 1.16/2.16.
[f] T(α) = T - T(SF) - T(IF).
[g] α = T(α)/T(SF).
[h] TRAT = T(α)/[T - T(α)].

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