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Summary

Conventional methods of nondestructive assay (NDA) have accurately assayed the plutonium content of many forms of relatively pure and homogeneous bulk items. However, physical and chemical heterogeneities and the high and variable impurity levels of many categories of processing scrap bias the conventional NDA results. The materials also present a significant challenge to the assignment of reference values to process materials for purposes of evaluating the NDA methods.

A recent study using impure, heterogeneous, pyrochemical residues from americium molten salt extraction (MSE) has been aimed at evaluating NDA assay methods based on conventional gamma-ray and neutron measurement techniques and enhanced with analyses designed to address the problems of heterogeneities and impurities. The study included a significant effort to obtain reference values for the MSE spent salts used in the study. Two of the improved NDA techniques, suitable for in-line assay of plutonium in bulk, show promise for timely in-process assays for one of the most difficult pyrochemical residues generated as well as for other impure heterogeneous scrap categories.

1. INTRODUCTION

Plutonium in the bulk forms generated by scrap recovery operations is often chemically impure and physically and chemically heterogeneous. The pyrochemical residues, in particular, are lean and highly impure chloride-salt-based materials in which the plutonium can coexist in both metallic and salt forms, the americium content is typically high (from a few to tens of weight percents relative to plutonium), and the typical bulk residue consists of heterogeneous and nonrepresentative chunks of various sizes. This describes the americium molten salt extraction (MSE) spent salts, in particular. Minimizing the handling of such highly radioactive materials requires assigning accountability values to these residues without removing them from the process line; they can then be immediately routed either to the next stage of processing or to waste disposal.

For the MSE spent salts (and for other pyrochemical residues) plutonium accountability values assigned by difference (residue value equals feed value minus product value) are grossly in error because they are the small difference of two large numbers, each with its own uncertainty. Routine crushing, blending, and sampling of combined MSE spent salt items for accountability is prohibited by the extreme inhomogeneities and high contact radiation dose rates. If calorimetric measurements, interpreted by gamma-ray isotopics for the bulk items, are used to obtain plutonium assays, pre-equilibration procedures must be incorporated as a result of the extremely long (12- to 20-h) thermal equilibration times required for the chunky residues. Moreover, when the americium content exceeds several percent (by weight relative to plutonium), even the state of the art in gamma-ray isotopics for heterogeneous bulk materials may not provide the required accuracy for the specific power (watts per gram of plutonium) because most of the heat is provided by the americium. Furthermore, the practicality of in-line calorimetry has not been demonstrated to date.

Segmented gamma-ray scanning (SGS) and passive neutron coincidence counting (PNCC) are two methods of measuring bulk plutonium that have been used in glove-box lines. Recent advances in plutonium nondestructive assays (NDAs) based on SGS[1,2] and

PNCC[3,4] show promise for the bulk assay of plutonium in MSE spent salts and other pyrochemical residues. A recent Los Alamos study[5] using MSE spent salts has evaluated these two new NDA approaches, in addition to the NDA of plutonium by calorimetry/gamma-ray isotopics. This study has two distinct phases: evaluating the NDA methods, and assigning reference values (for total plutonium and isotopic composition) to the 14 process residues (MSE spent salts) used in the study.

The first efforts to obtain reference values for the MSE spent salts involved completely dissolving the individual bulk items and sampling the dissolver for destructive analysis (DA).[5] This method was extremely difficult to control and was abandoned after it was applied to four of the residues used in the study. Subsequently, each remaining bulk residue was pulverized, sieved, blended, and sampled for DA. The metallic portion (remaining in the sieve) of each residue item was stoichiometrically oxided and reintroduced into the pulverized residue salt phase for blending. Because this second approach to obtaining reference values sacrifices only a few grams (out of 1 to 3 kg) of the bulk mass for DA, the method leaves behind homogenized reference materials with the bulk chemical composition of the process residues. It also provides an additional phase for the NDA evaluation in that each NDA method can be used again on the homogenized residues to independently evaluate the sensitivity of the method to the heterogeneities of the process materials.

A separate report on the details and results of the physical and chemical procedures used to characterize the residues is being prepared.[6] This report also includes the results of the evaluation of the NDA of the MSE spent salts by calorimetry/gamma-ray isotopics, and conclusions on its use as a reference technique for evaluating or verifying other NDA methods applied to these residues. Table I lists the bulk masses and the reference values for total plutonium mass and isotopic composition for each of the 14 MSE spent salts used in the study. The following sections describe the evaluations of the new SGS and PNCC methods for the NDA of spent MSE salts.

2. SGS ASSAYS WITH LUMP CORRECTIONS

2.1 Description of Method

The traditional SGS procedure was developed in the early 1970s.[7] In this procedure, transmission-corrected gamma-ray assays are performed on a rotating bulk item, segment by segment, to minimize some of the effects of heterogeneity. For each segment, the measured transmission is used to derive a self-attenuation correction. The assumption is that the bulk attenuation can be characterized by a single linear attenuation coefficient, which, in general, is

Table I. Reference Values for Total Plutonium and Isotopic Composition of the Original (Uncrushed) MSE Spent Salts

		Pu Weight Fractions from Chemistry						
Sample ID	*	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am (ppm)	Total Pu (g)
XBLP120	D	0.0151	93.732	5.9297	0.2754	0.048	26 081	112
XBLP267	D	0.013	93.872	5.9097	0.1842	0.021	35 142	126
XBLPs300	L D	0.013	93.642	6.0497	0.2473	0.048	3 036	199
XBLP270	D	0.009	93.992	5.8297	0.1506	0.019	33 737	99
XBLP121		0.0103	93.987	5.7256	0.2403	0.037	36 168	155.4
XBLP278		0.0077	94.599	5.2541	0.115	0.024	35 413	90.4
XBLPs301	L	0.0129	93.647	6.0431	0.247	0.05	2 519	247
RFMSE1		0.0067	93.973	5.853	0.147	0.02	53 633	243.8
RFMSE2		0.0101	93.805	5.9536	0.2032	0.028	47 482	372.7
RFMSE3	L	0.0097	93.826	5.86	0.2633	0.04	7 103	55.5
RFMSE4		0.0093	93.889	5.8719	0.2052	0.025	45 486	408.7
RFMSE5	L	0.0094	93.832	5.8576	0.2698	0.032	3 351	141.2
ARF595		0.0097	93.688	5.9354	0.3359	0.031	43 918	263.6
ARF642		0.0075	94.438	5.418	0.1213	0.015	45 777	219.5

*L = Lower Am content; D = Characterized following complete dissolution of bulk item.

dominated by the matrix. This procedure can provide unbiased assays if the item contains SNM in powders or fine particle sizes.

Unfortunately, much of the scrap and waste generated in the facilities contains SNM in the form of lumps, and if the SGS procedure is applied to these items, the assay results will be biased, usually in the negative direction. To minimize the bias caused by the lumps, lump-corrected segmented gamma-ray scanning (LCSGS) was developed. Details of the technique can be found in Ref. 1.

2.2 Comparison

The MSE spent salts are ideal for studying the differences between traditional SGS and LCSGS analyses because these salts are known to contain metallic lumps of plutonium. Fourteen MSE spent salt items were measured with the SGS, and the spectra for each item were analyzed by both methods. The traditional SGS assay results are summarized in Table II.

Table II. Summary of Traditional SGS Measurements of MSE Salt Samples

Sample ID	SGS (129) (g)	Sig (129) (g)	SGS (203) (g)	Sig (203) (g)	SGS (345) (g)	Sig (345) (g)	SGS (414) (g)	Sig (414) (g)
XBLP120	78.7	0.9	90.8	0.9	91.8	0.5	90.7	0.2
XBLP267	76.7	0.7	104.5	1.0	116.3	0.5	116.7	0.2
XBLPS300	108.2	0.8	147.6	0.9	164.0	0.4	163.9	0.2
XBLP270	60.5	0.6	81.5	0.8	89.8	0.4	89.7	0.2
XBLP121	83.9	0.6	116.7	1.2	127.0	0.8	128.6	0.5
XBLP278	52.8	0.5	68.3	0.7	73.3	0.4	74.0	0.2
XBLPS30[illegible]	92.7	0.9	134.2	1.2	179.2	0.6	186.1	0.3
RFMSE1	172.2	4.2	212.0	2.7	220.2	0.7	218.5	0.4
RFMSE2	149.4	14.6	241.5	0.6	322.6	0.8	326.3	0.7
RFMSE3	35.0	0.2	43.6	0.4	47.5	0.3	48.2	0.1
RFMSE4	156.6	7.7	262.1	5.0	357.2	0.9	358.6	0.7
RFMSE5	55.2	1.2	82.1	0.9	109.1	0.4	112.1	0.2
ARF595	176.7	14.3	227.8	7.5	248.1	0.6	244.2	0.3
ARF642	182.4	10.2	202.6	5.7	198.7	0.5	197.5	0.3

Notice that for some items there is a substantial increase in the SGS assay with increasing assay energy; this indicates that the items do contain SNM in lumps. The lump-corrected SGS assay results are summarized in Table III. The lump correction can be applied to the 414-keV assay, based on the difference between 129- and 414-keV assay results; these are listed under LCSGS(129) in the table. Similarly, the lump correction can be applied to the 414-keV assay based on the difference between 203- and 414-keV assays [listed under LCSGS(203)], as well as between 345- and 414-keV assays [listed under LCSGS(345)].

The lump correction can be applied in two ways: to the data, segment by segment, or to the entire item (the sum of all the segments). We found that the magnitude of the lump corrections for these MSE spent salts varies from segment to segment. The correction in this table has been made to the data from each segment individually. The comparison to the reference values is shown in Table IV and also in Fig. 1.

From this study, the following conclusion can be made. The traditional SGS assay is biased an average of 8.7% for these MSE spent salts. The LCSGS assay significantly reduces the bias from 8.7% to 4%. The remaining bias could arise from three sources. Some of these samples contain relatively large amounts of ^{237}Np, which emits a 415.76-keV

Table III. Lump-Corrected SGS Measurements of MSE Salt Samples

Sample ID	LCSGS (129) (g)	LCSGS (203) (g)	LCSGS (345) (g)	LCSGS Average (g)
XBLP120	93.4	94.0	91.5	92.97
XBLP267	124.5	123.5	119.4	122.47
XBLPS300	173.5	171.2	165.9	170.20
XBLP270	94.9	92.2	91.1	92.73
XBLP121	135.8	132.5	133.0	133.77
XBLP278	77.8	76.2	74.6	76.20
XBLPS301	206.6	214.0	209.2	209.93
RFMSE1	224.8	224.1	225.0	224.63
RFMSE2	353.5	358.3	342.8	351.53
RFMSE3	50.7	49.8	49.1	49.87
RFMSE4	394.4	399.2	373.5	389.03
RFMSE5	119.9	123.9	120.2	121.33
ARF595	252.5	250.5	251.5	251.50
ARF642	206.9	209.6	202.0	206.17

Table IV. Comparison of SGS Measurements with the Reference Values

Sample ID	Ref ^{239}Pu (g)	SGS (g)	LCSGS (g)	Lump Corr (%)	Ratio (SGS/Ref)	Ratio (LCSGS/Ref)
XBLP120	104.98	90.7	92.97	2.44	0.864	0.886
XBLP267	118.28	116.7	122.47	4.71	0.987	1.035
XBLPS300	186.35	163.9	170.20	3.70	0.880	0.913
XBLP270	93.05	89.7	92.20	2.71	0.964	0.991
XBLP121	146.03	128.6	133.77	3.86	0.881	0.916
XBLP278	85.51	74.0	76.20	2.89	0.865	0.891
XBLPS301	231.26	186.1	209.93	11.35	0.805	0.908
RFMSE1	229.13	218.5	224.63	2.73	0.954	0.980
RFMSE2	349.65	326.3	351.53	7.18	0.933	1.005
RFMSE3	52.13	48.2	49.87	3.35	0.925	0.957
RFMSE4	383.69	358.6	389.03	7.82	0.935	1.014
RFMSE5	132.50	112.1	121.33	7.61	0.846	0.916
ARF595	246.93	244.2	251.50	2.90	0.989	1.019
ARF642	207.29	197.5	206.17	4.21	0.953	0.995
				Average	0.913	0.959
				1σ	0.056	0.052

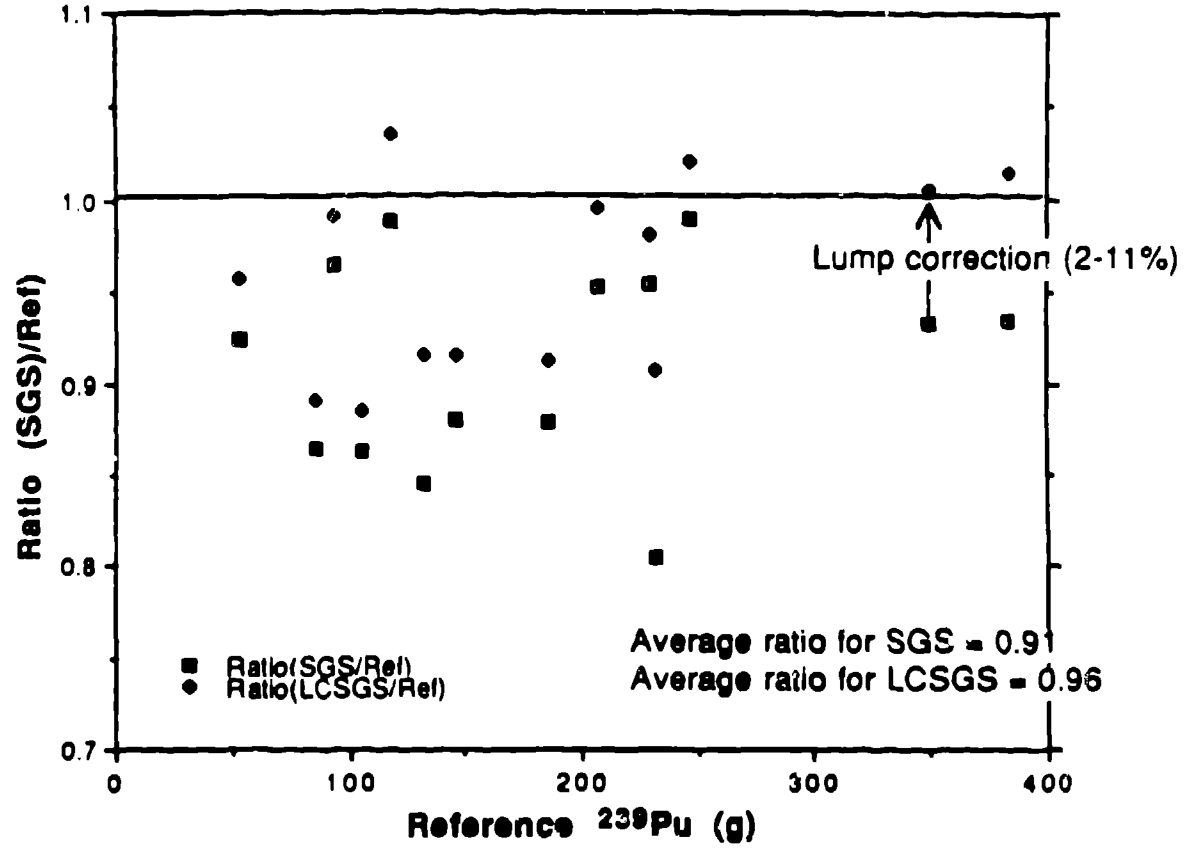

Fig. 1. SGS and LCSGS assays of the 14 MSE samples. The reference values are obtained by destructive analyses.

gamma ray through its ^{233}Pa daughter. This peak interferes with the 414-keV peak determination, and in the region-of-interest method of obtaining the peak area, the peak area is underestimated. Therefore the 414-keV assay could be biased low. A second possibility is a similar interference in the 414-keV background region caused by pulse pileup of the intense 208-keV gamma-ray photopeak. The third possible source of bias is that the LCSGS assumes that all the SNM lumps are the same size. If the SNM particle sizes are distributed, the correction may be biased, and the bias will depend on the particle size distribution.

3. PNCC MULTIPLICATION-EQUIVALENT-FISSILE (M-E-F) ASSAYS

3.1 Description of Method

The three unknown quantities in plutonium assays based on PNCC measurements are the mass of the plutonium, the neutron multiplication (M) of the unknown item, and the ratio (α) of α,n to spontaneous fission (SF) neutrons. For relatively pure materials, the value of α can be assigned based on the chemical composition so that the two measured quantities, the real coincidence (R), and total neutron (T) count rates can be used to solve both for M and for the effective ^{240}Pu mass, $^{240}\mathrm{Pu}_{\mathrm{eff}}$, the spontaneously fissioning component of the plutonium.[8] The known isotopic composition of the plutonium-bearing item is used to obtain the total plutonium mass from the assay result. Because impurities in the residue categories are large and variable, values for α cannot be assigned to residue materials. An alternate approach must be taken to perform the assay when α is unknown. The assays described use PNCC data obtained with the HLNCC-II.[9]

In the point model of passive neutron coincidence counting as described by Stewart,[10] T and R are expressed as α- and M dependent quantities. By substituting T into the equation for R to give

$$R = k_0(M^2)\,{}^{240}Pu_{eff} + k_1\,M(M-1)\,T \ , \tag{1}$$

the quantity α becomes included in the measured T. The constants in Eq. (1) can be defined, using Stewart's notation,[10] as

$$k_0 = \frac{\varepsilon^2 f}{2}\,\overline{\nu(\nu-1)}^{SF}\,n_{SF} \tag{2}$$

and

$$k_1 = \frac{\varepsilon f}{2}\,\frac{\overline{\nu(\nu-1)}^{(I)}}{\overline{\nu}^{(I)} - 1} \ , \tag{3}$$

where ε is the neutron detection efficiency, f is the fraction of neutrons counted in the coincidence gate, n_{SF} is the spontaneous-fission decay rate per unit mass of $^{240}Pu_{eff}$, and the quantities $\overline{\nu}$ and $\overline{\nu(\nu-1)}$ are the first and second moments (respectively) of the SF and induced (I) fission neutron multiplicity distributions. Note that for the HLNCC-II, k_0 is a well-established quantity ($18.14 \cdot s^{-1} \cdot g\ {}^{240}Pu_{eff}$) for assays of moisture-free bulk items.

The two unknowns in Eq. (1) can be reduced to one by describing the (unknown) M in terms of a multiplication-equivalent-(effective) fissile mass or M-E-F mass, $^{239}Pu_{eff}$, and a sample geometry factor, G:

$$M = \frac{k_2\,{}^{239}Pu_{eff}}{G} + 1 \ , \tag{4}$$

where k_2 is a constant and G is defined by the bulk shapes and dimensions of the unknown items. [For cylindrical packages of radius r and fill height h, G is equivalent to r(r + h)].[4] Similar to $^{240}Pu_{eff}$, the $^{239}Pu_{eff}$ is defined by the total plutonium mass and the isotopic composition of the plutonium-bearing item.[11] The ratio of the effective masses is γ such that

$$^{240}Pu_{eff} = \gamma \cdot {}^{239}Pu_{eff} \ , \tag{5}$$

where γ is determined by isotopic composition alone, independent of mass. Substitution of Eqs. (4) and (5) into Eq. (1) gives an equation in the measured quantities, R and T, with only one unknown, $^{239}Pu_{eff}$. In the limit, as (M - 1) approaches zero, appropriate for residues, the equation is simplified for two general cases: (i) very large α and (ii) undefined α.

3.2 Assay of M-E-F Mass When α is Very Large (Simple Self-Interrogation Approximation)

Substituting Eqs. (4) and (5) into Eq. (1) and rearranging gives

$$\frac{R_{IF}}{T} = A\left[\frac{^{239}Pu_{eff}}{G}\right]^2 + B\left[\frac{^{239}Pu_{eff}}{G}\right] + C \ , \tag{6}$$

where

$$R_{IF} = R - \gamma\,k_0\,{}^{240}Pu_{eff} \ , \tag{7}$$

$$A = k_1 k_2^2 , \tag{8}$$

and

$$B = k_1 k_2 . \tag{9}$$

For residues such as the first extraction MSE spent salts, $(M - 1)$ approaches zero, and the α values are very large because of the high (>1%) americium content. In this limit, the simple self-interrogation (SI) approximation is

$$C = \gamma k_0 (M^2 - 1)\, {}^{239}\mathrm{Pu}_{\mathrm{eff}} , \tag{10}$$

where C is shown[12] to be sufficiently small to be approximated by a constant in Eq. (6). The residues that qualify will have characteristically small values of R/T.

Calibration of the SI assay is performed with R and T data obtained from the high-americium crushed MSE spent salts by fitting a quadratic function to Eq. (6) to solve for A, B, and C. The results of this fit are shown in Fig. 2.

The assays of the original (uncrushed) MSE spent salts are based on a rearrangement of Eq. (6):

$$0 = a({}^{239}\mathrm{Pu}_{\mathrm{eff}})^2 + b({}^{239}\mathrm{Pu}_{\mathrm{eff}}) + c \tag{11}$$

where the coefficients,

$$a = \frac{AT}{G} , \tag{12}$$

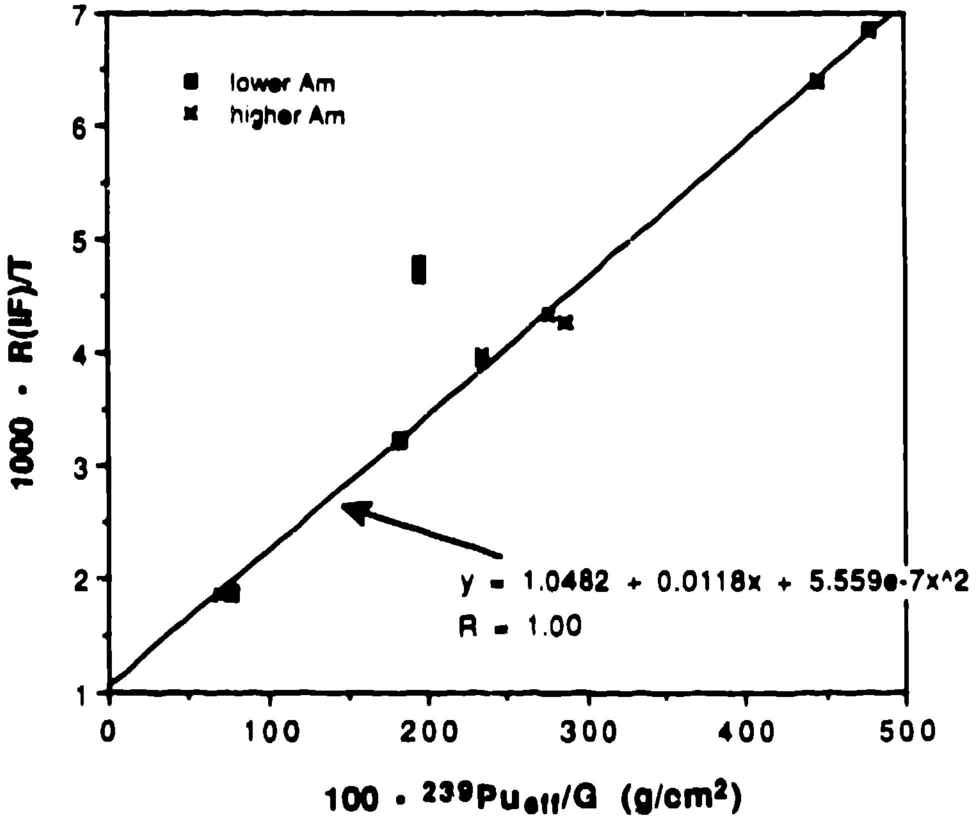

Fig. 2. Quadratic fit to crushed salt measurement results for calibration of SI assay.

$$b = \frac{BT}{G} + k_0 \gamma \ , \tag{13}$$

and

$$c = CT - R \ , \tag{14}$$

are unique to each unknown item. The SI assay result is

$$^{239}Pu_{eff} = \frac{-b + \sqrt{b^2 - 4ac}}{2a} \ . \tag{15}$$

The values of R, T, G, and γ are given elsewhere[4,12] for each MSE spent salt measurement (including multiple measurements of given salts in different containers corresponding to different G values). The ratio of the SI assay result to the reference value for each measurement is given in Table V. The average ratio is biased high if the (four) lower-americium items are included, consistent with expectations for these items. The ratios are also plotted vs $^{239}Pu_{eff}$ and vs percent americium in Fig. 3.

3.3 More General Assay of M-E-F Mass, Including Lower-α Cases

Some MSE spent salts resulting from a second extraction of americium from impure metal, and other plutonium residues with large but less than 1% americium content do not satisfy the criteria for SI. In this case, the measured ratio, R/T, is larger for a given residue category. One result of a decrease in α is that ε in k_1 [Eq. (3)] is no longer constant because the (generally higher energy) fission neutrons become a significant fraction of the neutrons produced. This limitation can be more significant for the chloride-based salts for which average α,n neutron energies are $\lesssim 1$ MeV compared to 2 MeV for fission neutrons. However, the americium content of the second extraction MSE spent salts is still relatively high ($\gtrsim 0.3\%$) so that typical α values are $\gtrsim 5$ for these materials. In these cases, the neutron energy spectrum is still dominated by α,n neutrons. Therefore, Eq. (1) is again used, assuming k_1 to be constant, with an approximation based on the limit as M - 1 approaches zero. The approximation includes

$$M^2 \cong 2M - 1 \tag{16}$$

and

$$M(M - 1) \cong M - 1 \ . \tag{17}$$

Substituting Eqs. (4), (5), (16), and (17) into Eq. (1) gives the M-E-F mass relationship to R and T that includes lower-α cases:

$$\frac{R}{T} = A'\left(\frac{^{239}Pu_{eff}}{G}\right)\left(\frac{^{240}Pu_{eff}}{T}\right) + B'\left(\frac{^{240}Pu_{eff}}{T}\right) + C'\left(\frac{^{239}Pu_{eff}}{G}\right) \ , \tag{18}$$

where

$$A' = 2k_0 k_2 \ , \tag{19}$$

$$B' = k_0 \tag{20}$$

and

$$C' = k_1 k_2 \ . \tag{21}$$

Table V. Comparison of M-E-F Assay Results with Reference Values

Sample ID	Reference $^{239}Pu_{eff}$ (g)	Can Type	$^{239}Pu_{eff}$/Reference from R(IF)/T vs $^{239}Pu/G$ (SI)	Can Average	$^{239}Pu_{eff}$/Reference from Small M Approximation (More General)	Can Average
XBLP120	110.5	#20	0.969	0.969	0.998	0.998
XBLP267	124.9	Tall	1.025		1.069	
		Tall	1.013	1.026	1.059	1.070
		Tall	1.040		1.083	
		#20	1.041	1.041	1.079	1.079
XBLPs300	193.6	Tall	1.047	1.047	0.977	0.977
		#20	1.083	1.083	1.021	1.021
XBLP270	98.1	Tall	1.068	1.068	1.080	1.080
		#20	1.116	1.116	1.126	1.126
XBLP121	153.9	Tall	1.010	1.002	1.040	1.032
		Tall	0.994		1.025	
		#20	1.068	1.077	1.088	1.097
		#20	1.087		1.106	
XBLP278	89.8	Tall	0.890	0.890	0.971	0.971
		#20	0.936		1.009	
		#20	0.959	0.949	1.030	1.021
		#20	0.951		1.023	
XBLPs301	240.3	Tall	1.095		1.030	
		Tall	1.099	1.108	1.034	1.043
		Tall	1.130		1.065	
		#20	1.147	1.153	1.108	1.115
		#20	1.159		1.121	
RFMSE1	244.3	P	0.929	0.933	0.936	0.940
		P	0.937		0.944	
RFMSE2	372.1	P	1.040	1.063	1.048	1.073
		P	1.086		1.098	
RFMSE3	54.2	P	1.003		0.937	
		P	1.021	1.018	0.953	0.950
		P	1.031		0.961	
RFMSE4	407.8	P	1.022	1.032	1.024	1.034
		P	1.042		1.045	
RFMSE5	137.4	P	1.084	1.019	1.047	1.022
		P	0.954		0.998	
ARF595	263.0	P	0.969	0.979	0.973	0.982
		P	0.988		0.991	
ARF642	218.8	P	0.926	0.935	0.937	0.946
		P	0.944		0.955	
		14 higher Am items average (1σ) =		1.006 (0.065)		1.032 (0.058)
		All 20 items average (1σ) =		1.025 (0.068)		1.029 (0.057)

Calibration for this more general approach that includes lower α values uses R and T data from all of the crushed MSE spent salts in a least squares fit to Eq. (18) to obtain A′, B′, and C′.

For assay of the original (uncrushed) MSE spent salts, Eq. (18) is rearranged to give

$$0 = a'\left({}^{239}Pu_{eff}\right)^2 + b'\left({}^{239}Pu_{eff}\right) + c' , \tag{22}$$

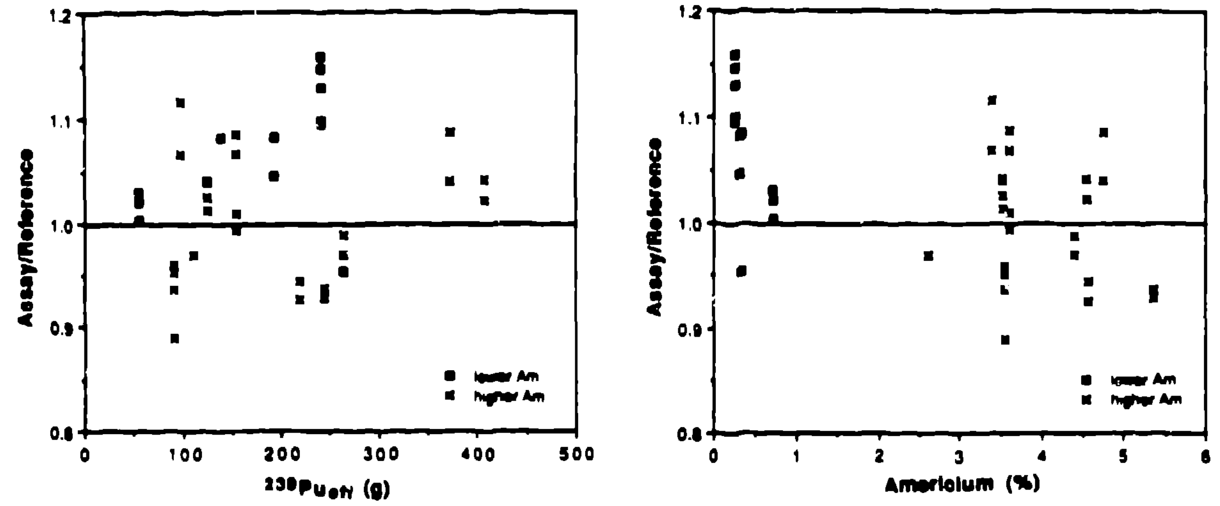

Fig. 3. The SI M-E-F assay results divided by the reference value vs $^{239}Pu_{eff}$ (left) and vs americium weight percent (right) for the original MSE spent salts.

where the coefficients,

$$a' = \frac{A'\gamma}{G} , \tag{23}$$

$$b' = B'\gamma + \frac{C'T}{G} , \tag{24}$$

and

$$c' = R \tag{25}$$

are unique to each unknown item. The assay result for this more general approach is

$$^{239}Pu_{eff} = \frac{-b' + \sqrt{b'^2 - 4a'c'}}{2a'} . \tag{26}$$

The ratio of the assay result to the reference value for each assay is given in Table V. A 3% bias appears in the average result, but the relative bias for the lower-americium MSE spent salts has vanished. The ratios are also plotted vs $^{239}Pu_{eff}$ and vs percent americium in Fig. 4.

4. CONCLUSIONS

Both the LCSGS and the M-E-F PNCC assays show promise for timely in-process assays of pyrochemical residues such as the MSE spent salts. The study demonstrates that 5% (1σ) assay results are achieved with the LCSGS. Further investigations will determine the cause of a 4% negative bias in the assay. Evaluation of the LCSGS results for the crushed MSE spent salts should help distinguish between ^{237}Np or 208-keV pileup interference and the variability of lump sizes as postulated causes of the bias. Elimination of the bias may also improve the assay uncertainty.

The M-E-F PNCC results obtained in the simple SI approximation are 7% (1σ) assay results, which are unbiased for those residues for which the americium content exceeds 1 wt% (relative to plutonium). A more general approach to the M-E-F PNCC assays eliminates the

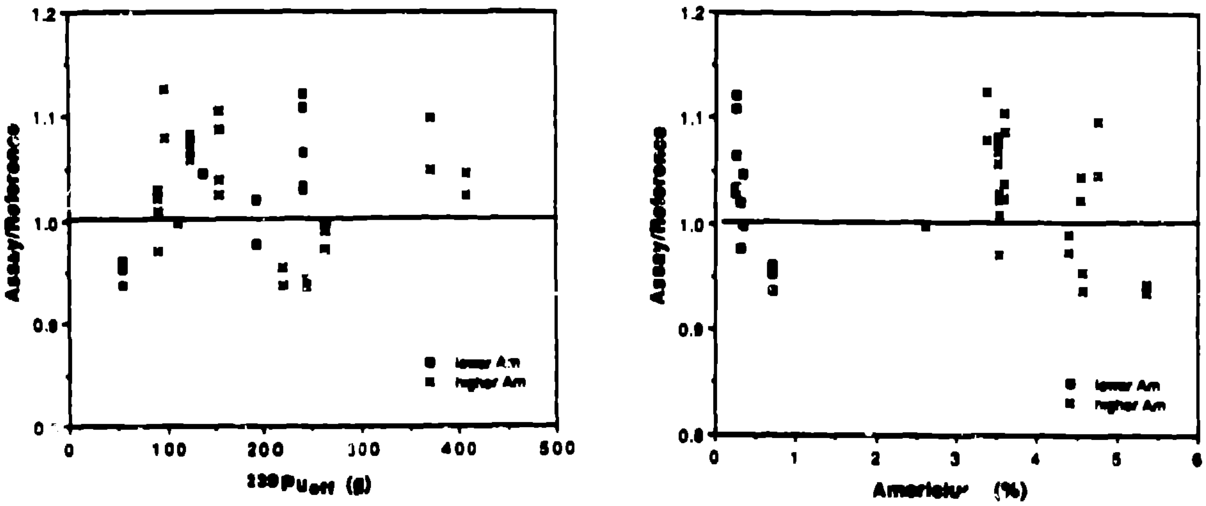

Fig. 4. The more general M-E-F assay results divided by the reference value vs $^{239}Pu_{eff}$ *(left) and vs americium weight percent (right) for the original MSE spent salts.*

positive bias that occurs in the SI results for the lower americium residues. The more general approach gives 6% (1σ) assays, but in this case the assays have a 3% positive bias. This bias may be caused by the energy-dependent neutron detection efficiency coupled with shifts in the neutron energy spectrum that result from variable α values and chemical differences in the matrix composition. Further investigations will include measurements designed to provide energy information that might be used to identify and correct these sources of bias. A longer term improvement is the use of a coincidence counter designed for minimum neutron energy dependence.

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