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Achieving Accurate Neutron-Multiplicity Analysis of Metals and Oxides with Weighted Point Model Equations

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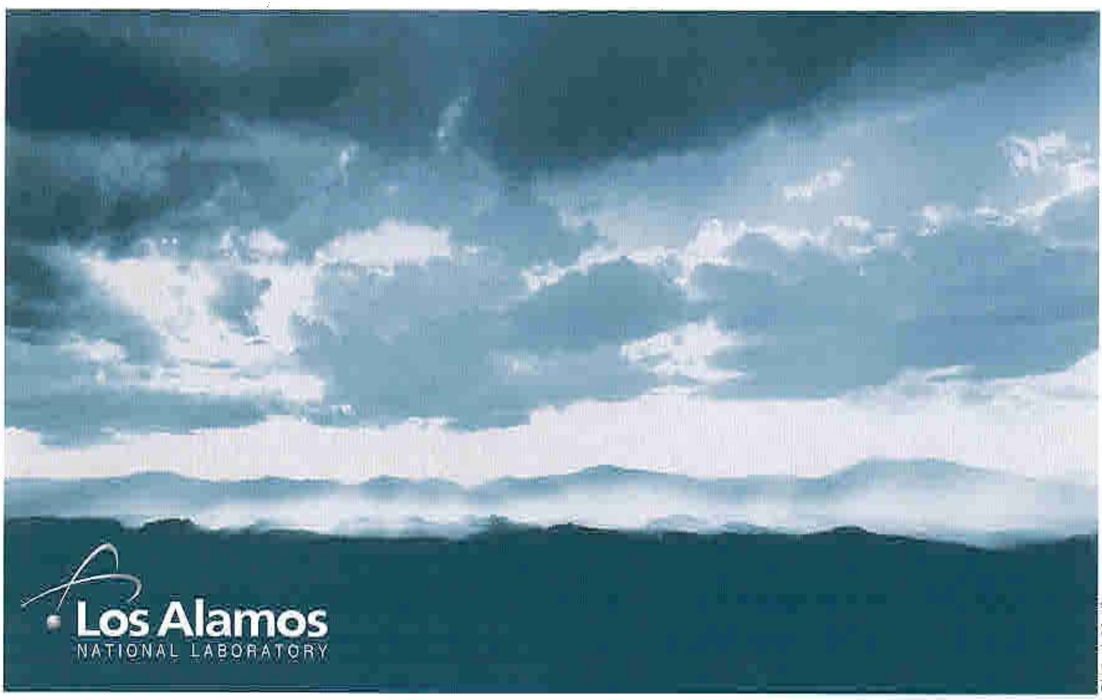

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ABSTRACT

Neutron multiplicity counting is a technique for the rapid, nondestructive measurement of plutonium mass in pure and impure materials. This technique is very powerful because it uses the measured coincidence count rates to determine the sample mass without requiring a set of representative standards for calibration. Interpreting measured singles, doubles, and triples count rates using the three-parameter standard point model accurately determines plutonium mass, neutron multiplication, and the ratio of (α,n) to spontaneous-fission neutrons (alpha) for oxides of moderate mass. However, underlying standard point model assumptions—including constant neutron energy and constant multiplication throughout the sample—cause significant biases for the mass, multiplication, and alpha in measurements of metal and large, dense oxides.

At Los Alamos National Laboratory a neutron- and photon-transport Monte Carlo code, MCNPX (Monte Carlo N-Particle eXtended) uses known physics processes and cross-section data to determine the neutron multiplicity distribution for material with user-specified composition and geometry. We vary material geometries, densities, and impurities in MCNPX and obtain simulated singles, doubles, and triples count rates. Weighting factors in the standard point model equations derived from these simulated count rates account for "non-point" behavior that causes biased measurements for the materials studied. We compare computationally predicted biases of the standard point model (difference between computational results for standard and weighted point model analyses) with those measured, and show the current status of this weighting technique for reducing bias in neutron multiplicity measurements. Potential safeguards advances are promising for both high-throughput accountability and receipts-verification for large numbers of items.

I. INTRODUCTION

Over the past twenty years thermal neutron multiplicity counting has become a standard nondestructive assay technique for impure plutonium materials [1]. In most applications the analysis is performed with the three-parameter point model multiplicity equations [2]; the neutron multiplication, alpha value [the ratio of (α,n) to spontaneous fission neutrons], and the effective ^{240}Pu mass [1] are calculated from the measured singles, doubles, and triples count rates corrected for dead time and background. The point model is based on a number of simplifying assumptions, the most important of which are the assumptions that (1) all neutrons have the same energy and (2) all neutrons have the same probability for inducing a fission (constant neutron multiplication). Neutrons from (α,n) reactions can have energies much different from fission neutrons, and this can lead to significant biases in the assay masses [3]. Neutron multiplication in plutonium metal samples and in high mass, high-density plutonium oxide samples can vary substantially throughout a sample and also lead to significant assay biases.

II. WEIGHTED POINT MODEL EQUATIONS

The weighted point model equations are included here from [4]. In this reference, the standard point model multiplicity equations were modified as follows:

$$S = mF_0 \varepsilon \nu_{s1} M(1+\alpha),$$
$$D = \frac{1}{2} mF_0 \varepsilon^2 \nu_{s2} f_d M^2 (f_D + \alpha f_D^{\alpha}),$$
$$T = \frac{1}{6} mF_0 \varepsilon^3 \nu_{s3} f_t M^3 (f_T + \alpha f_T^{\alpha}),$$

where

$$f_D = w_D[1 + a(M-1)],$$
$$f_D^{\alpha} = w_D^{\alpha} a(M-1),$$
$$f_T = w_T[1 + b(M-1) + c(M-1)^2],$$
$$f_T^{\alpha} = w_T^{\alpha}[d(M-1) + c(M-2)^2],$$

where

$$a = \frac{\nu_{s1}\nu_{i2}}{\nu_{s2}(\nu_{i1}-1)},$$
$$b = \frac{3\nu_{s2}\nu_{i2} + \nu_{s1}\nu_{i3}}{\nu_{s3}(\nu_{i1}-1)},$$
$$c = \frac{3\nu_{s1}\nu_{i2}^2}{\nu_{s3}(\nu_{i1}-1)^2},$$
$$d = \frac{\nu_{s1}\nu_{i3}}{\nu_{s3}(\nu_{i1}-1)},$$

where

$S, D, T =$ singles, doubles, and triples rates, respectively,
$m =$ effective ^{240}Pu mass,
$M =$ neutron multiplication,
$\alpha =$ ratio of (α,n) to spontaneous fission neutrons,
$F_0 =$ ^{240}Pu spontaneous fissions per gram per second,
$\varepsilon =$ neutron detection efficiency,
$f_d, f_t =$ doubles and triples gate fractions, respectively,
$\nu_{s1}, \nu_{s2}, \nu_{s3} =$ 1st, 2nd, and 3rd factorial moments of the ^{240}Pu spontaneous fission neutron distribution,

$\nu_{i1}, \nu_{i2}, \nu_{i3}$ = 1st, 2nd, and 3rd factorial moments of the neutron distribution from the neutron induced fission of ^{239}Pu, and where w_D, w_D^α, w_T, and w_T^α are the variable-multiplication weighting factors. The spontaneous fission and (α,n) contributions to the doubles and triples rates are all different functions of the multiplication and therefore have different weighting factors. The singles rate is proportional to the multiplication and so does not need weighting factors. When the four weighting factors are set to unity, the weighted point model equations become the standard point model equations.

III. THE WEIGHTING FACTORS

The triples weighting factor is shown in Figure 1 versus the leakage multiplication for plutonium oxide cylinders with masses from 1 to 6 kg, densities from 1.5 to 7.5 g/cc, and height/diameter ratios from 0.02 to 5.0. The source is an (α,n) source with a ^{240}Pu spontaneous fission neutron spectrum. The solid line is an unweighted least-squares fit to the data points. The dashed line is the equivalent weighting factor for Pu metals [4]. The other weighting factors show a similar relationship to the multiplication and have been fit.

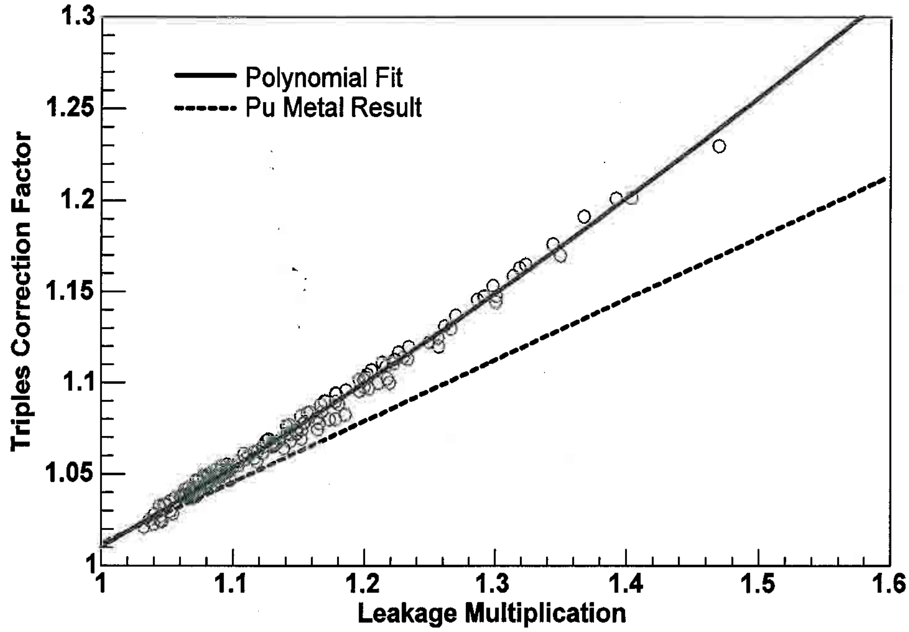

Fig. 1: Triples weighting factor versus leakage multiplication for all of the modeled plutonium oxide cylinders. The source is an (α,n) source with a ^{240}Pu spontaneous fission neutron spectrum. The solid line is an unweighted least-squares fit to the data points. The dashed line is the equivalent fit to the Pu metals [4], shown for comparison.

IV. WEIGHTED POINT MODEL SIMULATED ASSAYS

The assay to true effective ^{240}Pu mass ratio vs. the multiplication for the plutonium oxide cylinders is shown in Figure 2. The assay masses are calculated with both the standard (open symbols) and weighted (solid symbols) point model equations for an α value of 1. The average mass ratio from the weighted point model is 0.9993 ± 0.0002 with a standard deviation of 0.29% for these plutonium oxide samples. Also shown for comparison is the equivalent correction for Pu metal samples assayed with the standard point model (dashed line). Above about 1.3 in leakage multiplication, the corrections for oxides are comparable to the metals result. Below this value, the oxides have a smaller correction. There is also a slight (on the order of 1%) systematic rise with multiplication in the mass ratio. These systematic trends in the weighting technique are still under study.

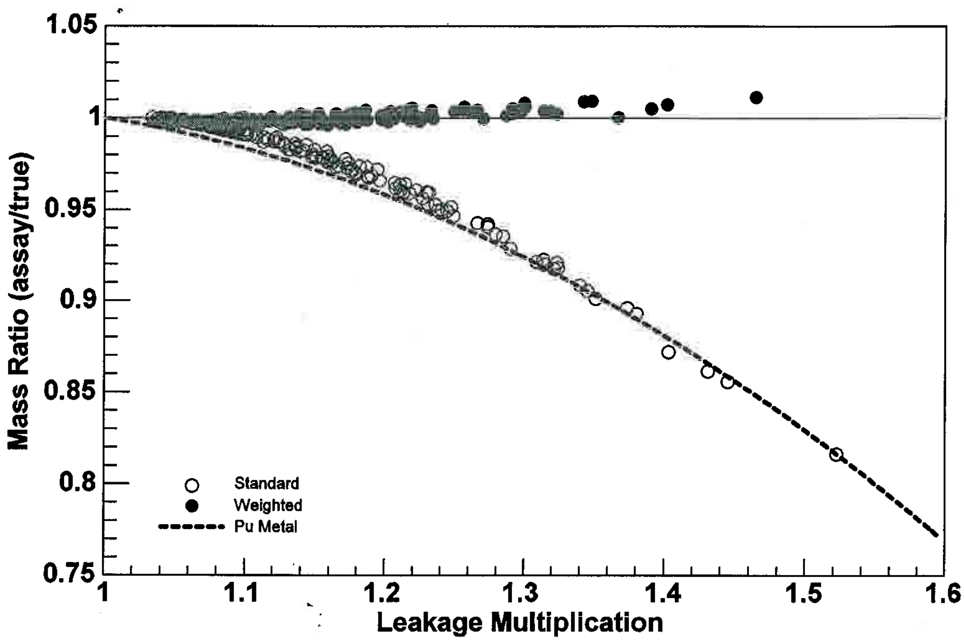

Fig. 2: Assay to true effective ^{240}Pu mass ratio versus leakage multiplication for the plutonium oxide cylinders that were used to determine the doubles and triples weighting factors. The assay masses are calculated with both the standard (open symbols) and weighted (closed symbols) point models for an α value of 1. The dashed line corresponds to the equivalent ratio for Pu metal samples with the standard point model.

To test the weighting factors, an additional 25 random samples of plutonium oxide were generated, as listed in Table I. For these 25 samples, the mass, density, diameter, and alpha values were all randomly selected within the same ranges used to determined the weighting factors.

Table I. *Data for 25 randomized samples of plutonium oxide.*

Sample Number	PuO_2 Mass (kg)	Density (g/cc)	Height/Diameter (cm/cm)	Alpha value
1	5.8	2.2	0.49	0.37
2	1.1	5.0	0.13	0.38
3	5.8	3.4	0.66	1.2
4	2.7	2.4	0.81	0.43
5	3.6	3.3	0.41	2.7
6	3.6	1.7	0.33	1.5
7	4.3	2.4	1.0	0.41
8	3.3	7.2	0.09	0.93
9	5.8	4.4	0.48	0.55
10	5.6	5.0	0.44	0.65
11	3.6	7.1	0.20	2.7
12	4.1	2.9	1.3	1.4
13	5.7	6.5	0.18	1.4
14	3.2	3.4	0.16	1.4
15	3.0	4.3	0.20	0.77
16	5.6	4.4	0.35	0.50
17	4.4	7.4	0.18	2.7
18	3.3	3.0	0.51	1.3
19	3.6	5.9	0.13	1.3
20	2.4	4.3	0.21	0.68
21	3.6	2.6	0.43	0.62
22	2.1	4.7	0.29	0.64
23	5.3	3.3	0.51	1.6
24	1.4	7.3	0.036	0.95
25	2.8	2.8	0.16	1.3

Analyzing the Monte Carlo calculations with the standard point model tended to overpredict the multiplication and alpha value while underpredicting the mass. This is the same trend that was seen in the metal study [4]. The assay mass over the true mass for the 25 randomized samples is shown in Figure 3. The average assay/true mass ratio for the weighted model is 1.00 ± 0.04 with a standard deviation of 0.2%. The results were found to be biased by as much as 5% using the standard point model. The points for the standard point model calculations show a lot of scatter in addition to the bias because of the randomized density and alpha values.

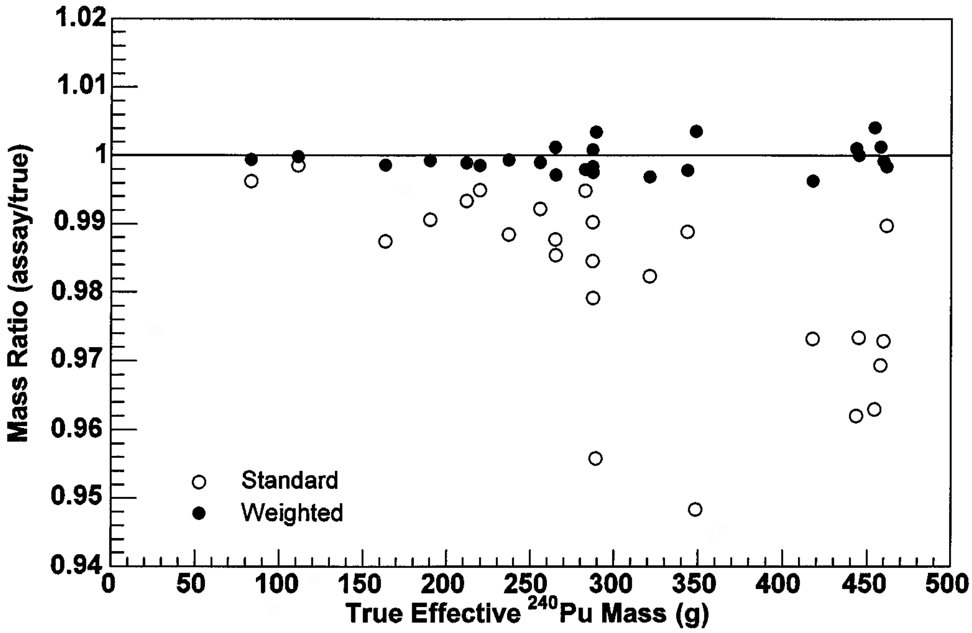

Fig. 3: Assay to true mass ratio vs. true effective ^{240}Pu mass for 25 randomized plutonium oxide samples. The calculations are done with both the standard (open points) and weighted (closed points) point models.

V. SPECIAL ASSAY CONSIDERATIONS

All of the Monte Carlo calculations used to determine the weighting factors assumed a low burn up isotopic composition listed in Table II. To see if the isotopic composition affects the weighting factors, additional Monte Carlo calculations were done with the isotopic composition for ARIES materials listed in Table II. The mass correction factor for a 4-kg oxide with density 6 g/cc and diameter 14 cm is 3.9% using the ARIES isotopic composition but decreases to 2% using the isotopic composition for the weighting factors. It is important to know the isotope composition of the material that will be assayed in order to obtain the most suitable weighting factors.

Table II. *Two different isotopic compositions used in the Monte Carlo calculations.*

Pu Isotopic Composition	Weighting Factors	ARIES Materials
^{238}Pu	0.013	--
^{239}Pu	0.595	0.940
^{240}Pu	0.238	0.060
^{241}Pu	0.104	--
^{242}Pu	0.050	--

VI. COMPARISON WITH EXPERIMENT

A comparison of the two analysis methods was made with experimental data with six plutonium oxide standards assayed in the ARIES detector [5]. The isotopic compositions are known for these samples from destructive analysis. For the weighted point model the count rates were analyzed using the Monte Carlo determined weighting factors. In Table III, the ARIES measurements are shown with and without the weighting method. Using the standard point model equations, the bias in the mass ratio (declared mass, or "true" mass, divided by the assay mass) varies between 1.9 to

8.4%. The weighted point model resulted in better assay values for all the samples except for LARI-MC-0006. The results are also displayed in Figure 4, where the open symbols are the standard mass ratios and the solid are the weighted mass ratios. The red arrow indicates the known systematic uncertainty due to the incorrect isotopic composition used to determine the weighting factors (assuming the ARIES isotopic composition as discussed in Section V).

Table III. *ARIES measurements with and without the weighting method. The weighting factors assume a nominal isotopic composition of 94% ^{239}Pu.*

Source	Plutonium Mass (g)	Alpha	Multiplication	Standard Mass Ratio (Declared/Assay)	Weighted Mass Ratio (Declared/Assay)
LARI-MC-0002	100	1.4	1.00	1.019 ± 0.005	1.023 ± 0.005
Calex 1	398	0.985	1.10	1.027 ± 0.007	1.021 ± 0.007
LARI-MC-0003	750	0.806	1.13	1.032 ± 0.007	1.018 ± 0.007
LARI-MC-0004	1500	0.926	1.21	1.04 ± 0.01	1.00 ± 0.01
LARI-MC-0005	3000	0.806	1.34	1.07 ± 0.01	0.98 ± 0.01
LARI-MC-0006	4433	1.08	1.48	1.084 ± 0.006	0.915 ± 0.006

The high alpha value for the first sample, LARI-MC-0002, can be explained because it consists of a matrix of diatomaceous earth whereas all others are oxide. Systematic uncertainties observed in the weighted point model analysis would result in an over correction of the assay mass. As observed in Figure 2, there is a slight systematic rise (on the order of about 1% in increase from 1) with multiplication in the corrections. As discussed in the previous sections, the corrections using the weighted point model overcorrect the ARIES data because the isotopic are different than what was used to determine the weighting factors. These reasons still do not account for magnitude of the overcorrection of the oxide sample LARI-MC-0006. One difference of the oxide LARI-MC-0006 from the other oxide samples was that it was made from a different batch of material. At the time of this study, the value of 0.915 for the last sample is still not understood.

VII. CONCLUSIONS

Many items with varied material geometries, densities, and impurities were modeled with MCNPX to obtain simulated singles, doubles, and triples count rates. Weighting factors in the standard point model equations derived from these simulated count rates account for "non-point" behavior that causes biased measurements for the materials studied. We compared computationally predicted biases of the standard point model (difference between computational results for standard and weighted point model analyses) with those measured in the ARIES detector. Using an assumed isotopic composition, the weighting technique improves the bias of the ARIES mass assays to approximately 2%, an improvement from the 2–7% variation obtained using the standard equations alone. Improvements in the weighting technique include using a plutonium isotopic composition that is closer to the ARIES data in the MCNPX calculations and increasing the collection of samples that have a larger leakage multiplication. The current status of this weighting technique for large samples of plutonium oxides shows promise.

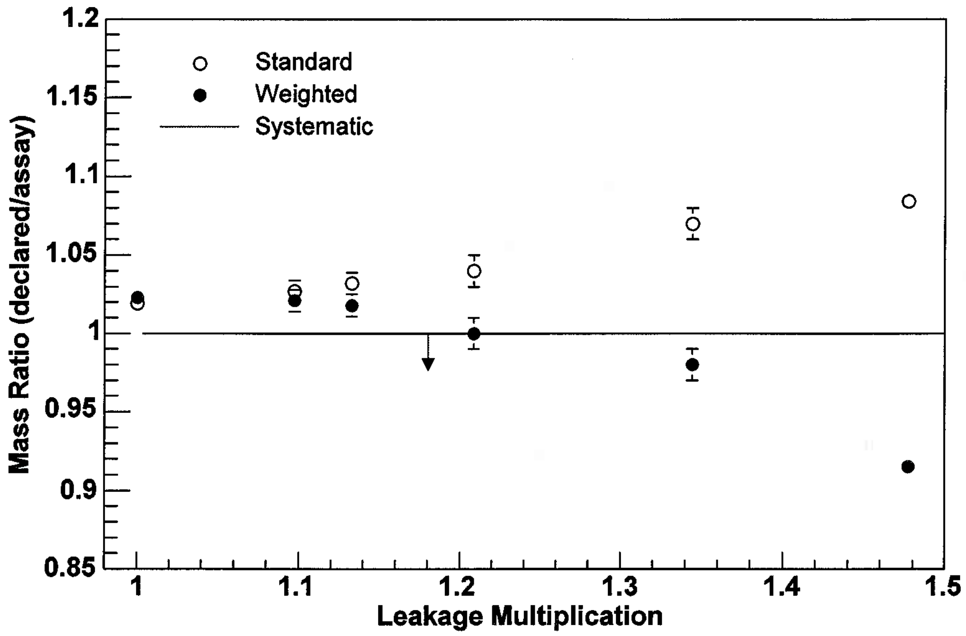

Fig. 4. *Shown is the mass ratio versus multiplication for six plutonium oxide standards assayed using the ARIES detector [5] and analyzed with the standard (open symbols) and weighted (solid symbols) point model equations. The red arrow indicates the known systematic uncertainty in the weighting factors based on determining the weighting factors with a different isotopic composition for plutonium as discussed in Section V. For the sample with the highest leakage multiplication, see discussion in the text.*

VIII. ACKNOWLEDGEMENTS

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