EXPECTED PRECISION FOR NEUTRON MULTIPLICITY ASSAY USING HIGHER ORDER MOMENTS

N. Ensslin, A. Gavron, W. C. Harker, M. S. Krick, D. G. Langner, M. C. Miller, and M. A. Pickrell
Los Alamos National Laboratory
Safeguards Science and Technology Group, NIS-5
Los Alamos, NM 87545 USA

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Abstract

This paper reports on the development of a new Figure of Merit code that can calculate the expected precision in neutron multiplicity assay using higher order moments. The code is used to provide a first look at the quadruple coincidence count rate and its expected precision. The results are good enough to warrant further study of potential applications of quadruple (quad) coincidences for large multiplying plutonium items. Also, the new code makes it possible to estimate the multiplicity assay precision if only randomly-triggered moments are used. This approach is described briefly, along with the current status of the investigation.

Introduction

Passive neutron multiplicity counting is coming into use in Department of Energy facilities to improve the assay of bulk, impure plutonium metals, oxides, scrap, and residues. Three measured parameters—single, double, and triple neutron events—are used to solve for sample $^{240}$Pu effective mass, self-multiplication M, and $(\alpha,\gamma)$ reaction rate $\alpha$. The solution equations are based on the point model developed by Boehnel (Ref. 1) and Hage and Cifarelli (Ref. 2 and 3). This model treats the sample as a single point in space to relate the moments of the measured and calculated multiplicity distributions.

In recent years there has been some curiosity about the next higher moment, representing quadruple (quad) neutron coincidence events. When modern, high-efficiency multiplicity counters are used to assay large plutonium (Pu) metal or oxide items, the triple coincidence rate can be comparable to or even larger than the double coincidence rate. This suggests that the rate of quad coincidences could also be large enough to measure with useful statistical precision. Also, recent data on bulk Pu samples, such as that collected at Lawrence Livermore National Laboratory (Ref. 4), show that the three-parameter point model does not always describe the sample accurately enough. This suggests that the use of quad coincidences could potentially improve assay accuracy under some circumstances. For example, could the quads/triples ratio provide a correction for nonuniform multiplication (i.e., variable neutron worth) in large Pu metal items?

Most commercially-available multiplicity electronics packages measure both signal-triggered multiplicity distributions (also called foreground, or "R+A") and random-triggered multiplicity distributions (also called background, or "A"). From these distributions, the software analysis package usually extracts the correlated moments of the emitted multiplicity distribution by unfolding the background moments from the foreground moments. Although not widely known, it is also possible to extract the correlated moments solely from the measured background moments by using moments of one higher order. The mathematics for this approach is given by Hage and Cifarelli (Ref. 2). Also, this approach has been implemented for some time in electronics circuits based on the "Feynman Variance" approach (Ref. 5).

To investigate the potential usefulness of quad coincidences, we have developed a new Figure of Merit code that estimates the expected count rate and precision for events up to the fourth order. The

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The new code also provides the higher moments needed to estimate the expected precision for multiplicity assay using randomly triggered distributions. This paper outlines the analysis procedure, the process used to benchmark the code, the results to date for quads and randomly triggered multiplicity analysis, and some potential future applications.

**Figure of Merit Estimation Procedure**

The new Figure of Merit code used in this paper is based on an earlier code developed to support the neutron multiplicity counter design process and to predict the expected performance (Ref. 6). The new code predicts the expected single, double, triple, and quad count rates and their estimated precision by calculating the factorial moments of the expected neutron multiplicity distribution. The multiplicity distribution does not need to be measured, but is predicted from preselected sample and detector design parameters. The code then predicts the error in the multiplicity assay using either foreground-minus-background moments or only background moments.

The new code features a completely reorganized analytical section to make the extension to higher moments more transparent. Some errors in the old code have been corrected, but they were not large enough to significantly affect previously reported results. The subtraction of random background events has been improved, and errors in the previous treatment of binomial correlations have been removed. However, comparisons with Monte Carlo simulations and measured data (Sections III and IV below) suggest that further improvements in the treatment of correlations between events may be needed.

The analytical procedure used in the code can be outlined as follows. A more detailed derivation will be published separately.

1. Input to the code consists of detector parameters such as efficiency, die-away time, gate length, predelay, count time, and background rate. Sample $^{240}$Pu effective mass, $M$, and $\alpha$ must also be entered.

2. From the detector and sample input parameters, and the known nuclear data for the spontaneous and induced fission factorial moments in $^{240}$Pu and $^{239}$Pu, the code computes the expected superfission factorial moments $v_1$ through $v_7$. The procedure for deriving these moments from probability-generating functions is given by Boehnel in Ref. 1, and the first three moments are listed in Ref. 6 in the format used by the code.

3. The factorial moments $r_1$ through $r_6$ of the signal-triggered distribution are computed from $v_2$ through $v_7$, and the factorial moments $s_1$ through $s_6$ of the random-triggered distribution are computed from $v_1$ through $v_6$. The necessary relationships are given in Ref. 2.

4. The measured background moments $b_1$ through $b_6$ are computed from $s_1$ through $s_6$. Then the factorial moments $f_1$ through $f_6$ of the measured foreground distribution can be unfolded from $r_1$ through $r_6$ and $b_1$ through $b_6$. The underlying equations are given in Ref. 2.

5. The double, triple, and quad coincidence count rates (with room background removed) are defined as

$$D = Sr_1,$$  \hspace{1cm} (1)

$$T = Sr_2/2,$$  \hspace{1cm} (2)

$$Q = Sr_3/6.$$  \hspace{1cm} (3)
Fig. 1. Double, triple, and quad coincidence count rates as a function of $^{240}\text{Pu}$ effective mass for a neutron multiplicity counter with detection efficiency = 50%, die-away time = 50 microseconds, gate width = 64 microseconds, predelay = 3 microseconds, and background = 10 counts/s. The solid lines are for multiplication values typical of Pu metal, and the dashed lines are for Pu oxide with $\alpha = 1$.

Figure 2 illustrates the relative standard deviation (RSD) for the triple and quad coincidence rates, $dT$ and $dQ$, as obtained from the Figure of Merit code for the same conditions as in Figure 1, with 1000-s counting time. For bulk Pu oxide, the RSD in the quads is in the range of 4 to 7% for $\alpha = 1$, and in the range of 10% to 30% for $\alpha = 5$ (not illustrated). For the same measurement parameters, the RSD in the multiplicity assay of Pu oxide using the singles, doubles, and triples is in the range of 0.5% to 3% (see Fig. 3 below). This implies that quad coincidence information is not likely to improve the multiplicity assay of Pu oxide.

For bulk Pu metal, the RSD in the quads is in the range of 0.5% to 2% for $\alpha = 0$ to 5. This is comparable to the RSD in the multiplicity assay of Pu metal and residues, and implies that quad coincidence information might improve assay results. However, this may depend on whether the RSD obtained from the Monte Carlo Neutron Photon (MCNP) simulation, which is roughly 4 times larger, is more correct than that obtained from the Figure of Merit code. Also, it would be essential to develop an accurate deadtime correction procedure before a correct, quantitative value for the quads could be obtained.
Fig. 2. Estimated relative standard deviation (RSD) for the triple and quad coincidence count rates as a function of $^{240}$Pu effective mass for the multiplicity counter described in Figure 1, with 1000s counting time. The solid lines are for Pu metal, the dashed lines are for Pu oxide. For Pu metal, the quads RSD is shown as a broad band, the reflect the range of uncertainty based on the current status of the benchmarking process.

Fig. 3. Comparison of multiplicity assay precision using signal-triggered foreground-minus-background moments (solid line) versus using only random-triggered background moments (dashed line) for 1000s assay of Pu oxide ($\alpha = 1$) using a neutron multiplicity counter with detection efficiency = 50%, die-away time = 50 microseconds, gate width = 64 microseconds, predelay = 3 microseconds, and background = 10 counts/s.
6. In terms of the measured foreground and background moments,

\[ D = S(f_1 - b_1), \]  
\[ T = S(f_2 - b_2 - 2b_1(f_1 - b_1))/2, \text{ and} \]
\[ Q = S[f_3 - b_3 - 3b_2(f_1 - b_1) - 3b_1(f_2 - b_2 - b_1(f_1 - b_1))]/6. \]

7. Or, using only the measured background moments,

\[ D = S(G, ...)\left( b_2 - b_1^2 \right), \]  
\[ T = S(G, ...)\left( b_3 - 3b_2b_2 + 2b_1^3 \right), \text{ and} \]
\[ Q = S(G, ...)\left( b_4 - 4b_1b_3 + 12b_2b_2 - 6b_1^4 - 3b_2^2 \right). \]

The expressions \((G, ...)\) refer to three different normalization coefficients composed of appropriate combinations of gate fractions, die-away time, efficiency, and other constants (Ref. 6). Note that Eq. 7, 8, and 9 for \(D, T,\) and \(Q\) each require one higher moment that Eq. 4, 5, and 6.

8. The error in the quads \(dQ\) is obtained from a procedure similar to that described in Ref. 6, but factorial moments up to \(f_6\), which involves \(v_7\), are utilized.

9. The error in the triples \(dT\), needed to compute the error in the foreground-minus-background multiplicity assay, uses moments up to \(f_4\), which involves \(v_5\). When only background-triggered moments are used, the error in the triples uses moments up to \(b_6\), which involves \(v_6\). (Note that the error in a multiplicity distribution of order \(N\) uses moments up to order \(2N\), because the variance is computed by combining multiplicity distributions in quadrature.)

10. If the ratio \(Q/T\) is used as a correction factor, the covariance \(\text{cov}(T,Q)\) between \(T\) and \(Q\) is needed. This also requires moments up to \(f_5\), which involves \(v_6\).

11. Finally, the procedure for determining the multiplicity assay error utilizes the error in \(D\) and \(T\), the covariance between \(D\) and \(T\), and the derivatives of the sample's fission rate with respect to \(D\) and \(T\), as detailed in Ref. 6.

Comparison with Monte Carlo Simulation

The new Figure of Merit code was benchmarked in two different ways, by comparison with Monte Carlo simulation and by comparison with measurements of californium neutron sources. This section describes the comparison with Monte Carlo simulation of neutron detection and multiplicity electronics data processing.

The simulation code specifies the detector and sample properties in much the same way as the Figure of Merit code. Then, a random number generator is used to select the spontaneous fission multiplicity and the probability of induced fission in each generation of spontaneous fission or \((\alpha,n)\) neutrons. This allows for a realistic simulation of sample self-multiplication. The code yielded values for
quadruples that were about 10% higher than the Figure of Merit code, with the triples and doubles in even closer agreement.

If the Monte Carlo code is run repeatedly with different starting values for the random number generator, it is also possible to determine the standard deviations in the count rates. A comparison with the Figure of Merit code at low multiplication yielded roughly 50% higher standard deviations in the triples and quads, but a comparison at high multiplication yielded roughly 4 times higher standard deviations in the triples and quads. This difference is still under investigation, but the range of uncertainty between the two approaches is reflected in the broad band shown in Fig. 2 below.

**Comparison With Multiplicity Counter Data**

We compared calculated and observed quadruple standard deviations for a series of californium sources ranging in strength from 1,000 to 12,000 n/s, using both a 29%-efficient waste drum counter and a 53%-efficient 5-ring multiplicity counter. The standard Los Alamos Neutron Coincidence Code (NCC) was modified to collect quad coincidences and determine the standard deviation in long series of 1000-s runs. The Figure of Merit code was set up to use the actual doubles and triples gate fractions for each counter, but the quads gate fraction was estimated from the other two gate fractions. For the californium measurements, the code was adapted to use the spontaneous fission nuclear data constants for californium, self-multiplication was set to 1, and α was set to 0.

The measured californium quad count rates were about 10% higher than those calculated from the Figure of Merit code. The measured standard deviations were about 35% higher than calculated. In general, both the californium measurements and the Monte Carlo simulations yield quad rates and standard deviations that are consistent with each other at low multiplication, and that are somewhat higher than the estimates from the Figure of Merit code. These differences are similar to those found in past benchmarking studies and are attributed to the fact that the computation of variance from the calculated multiplicity distributions does not completely include the effect of correlations between events.

Additional benchmarking studies will be carried out with bulk plutonium samples to help understand the effect of multiplication on the standard deviation in the quad count rate. Also, more benchmarking studies are needed for the variance in randomly triggered distributions.

**Results For Quad Coincidences**

Figure 1 illustrates quad coincidence count rates calculated with the new Figure of Merit code for a 50%-efficient neutron multiplicity counter. By way of comparison, the associated double and triple coincidence count rates are also plotted. Figure 1 covers two important classes of bulk plutonium: the solid lines are for multiplication typical of metal (M = 1.5 and α = 0 at 60 g $^{240}$Pu effective), and the dashed lines are for oxide (M = 1.075 and α = 1 at 60 g $^{240}$Pu effective).

Figure 1 demonstrates that the quadruple count rates are very low for small Pu samples and would not be useful for waste assay. However, the quadruple rates rise rapidly with increasing mass. With a modern, high-efficiency multiplicity counter, the quadruple rates are easily measurable for bulk plutonium oxide, scrap, or residues. For kilogram quantities of plutonium metal, such as 1 kg of metal with 6% $^{240}$Pu, the quadruples can be higher than the triples, which are in turn higher than the doubles.
Quads/Triples Correction Factor

Given the relatively large uncertainty in the quad coincidence RSD, and the lack of a deadtime correction for quads, it is unlikely that quads can be used directly for assay. It would certainly be very complicated analytically to incorporate the quads directly into the present multiplicity analysis, which already requires the solution of a cubic equation for multiplication in terms of the triples/doubles ratio. Instead, it may be better to define a correction factor (CF) based on the quads/triples ratio, such as (corrected multiplicity assay) = (initial multiplicity assay) x CF, where

\[ CF = 1 + kQ/T \]  

(10)

In this way, the existing multiplicity assay approach is unchanged, and its usually good RSD is less affected by the relatively poor RSD in the quads. The errors would be propagated as follows:

\[
\frac{d(\text{corrected assay})^2}{(\text{corrected assay})^2} = \frac{d(\text{initial assay})^2}{(\text{initial assay})^2} + \frac{dCF^2}{CF^2} \]

(11)

We can derive that the error in the correction factor would be

\[
dCF = \frac{k}{T}((dQ)^2 + (QdT/T)^2 - 2Q\text{cov}(T,Q)/T))^{1/2} \]

(12)

The relative error in the correction factor would be

\[
\frac{dCF}{CF} = \frac{kQ}{T} \frac{dQ}{Q} \]

(13)

When the relative error in the correction factor is written in this form, we can see that it may yield a relatively small incremental error, depending on the application.

In Ref. 4, it was shown that variations in multiplication or neutron worth within large or distributed Pu metal items can affect the multiplicity assay by 5% to 20%, depending on mass and geometry. From Ref. 4 we can estimate that the coefficient k in Eq. 13 is approximately 0.049. Under these conditions, dCF/CF may be much less than 1% even for relatively large values of dQ/Q, or even if Q is comparable to T. However, note that a relatively small error is only one necessary condition on the use of CF. It has not yet been determined if Q/T has a meaningful physical basis as a correction factor, or if an accurate deadtime correction for Q can be obtained.

Preliminary Results Using Random-Triggered Moments

Figure 3 illustrates the first preliminary comparison of the RSD obtained from signal-triggered foreground-minus-background moments (Eq. 4 through 6) versus the RSD from randomly triggered background moments only (Eq. 7 through 9). The Figure of Merit code was used under the following conditions: 1000-s total counting time; total number of random-triggers set equal to the count time divided by the gate width; foreground and background gate widths both set to 64 µs. These conditions have not yet been varied to obtain optimum, operating parameters. Also, we do not know what deadtime corrections are required for randomly triggered moments, or how sensitive any calibration procedure would be to the choice of the "appropriate constants" mentioned in Eq. 7 through 9.
For the calculations shown in Fig. 3, the singles, doubles, and triples count rates are identical for both approaches. This follows from the use of Eq. 1 through 3 to define the correlated moments. Also, the final assay results are computed from the singles, doubles, and triples in the same manner as before (Ref. 6) for both approaches. However, Fig. 3 suggests that the RSD can be different, and that in some circumstances the RSD may be slightly lower if randomly triggered moments are used. These preliminary results are intriguing, but given the very serious caveats outlined in the previous paragraph, it is clear that the use of randomly triggered moments requires much more study.

Potential Applications

If the initial results for quad coincidences presented in this paper are validated, and if a quantitative deadtime correction for quads can be developed, there are several potential applications. Reference 4 and other publications have shown that the current three-parameter multiplicity analysis procedure, based on the point model developed by Boehnel (Ref. 1) and Hage and Cifarelli (Ref. 2, 3) is not exact for large, distributed, multiplying Pu items. Quad coincidences may provide a correction factor for metal items with variable multiplication or for other point model deficiencies. Also, quads may provide a fourth neutron coincidence attribute for bilateral or trilateral inspections of excess weapons materials.

For randomly triggered multiplicity analysis, if the very preliminary results for the RSD are validated, and if an appropriate deadtime correction for the third-order background moment can be developed, there may be an application for this approach. Wherever the overall assay precision or count time could be reduced by even a small factor, DOE facility throughput on accountability measurements or excess weapons inspections could be increased, resulting in time and cost savings.

Conclusions

This paper is the first attempt to look at quad coincidences, and much remains to be done. These conclusions summarize the current status of the work.

1. For bulk plutonium oxide and metal samples, the quad coincidence rate in a high-efficiency multiplicity counter is high enough to be easily measured.

2. The calculated RSD for quads appears to be good enough for use with plutonium metal samples, but not with oxide samples. Current differences between the Monte Carlo and Figure of Merit results need to be resolved.

3. A quads/triples correction factor can be used to avoid a large increase in multiplicity assay RSD. However, a physical basis for such a correction factor has not yet been established.

4. A deadtime correction procedure for the quad moment needs to be developed before quads, or quads/triples, can be used in a quantitative fashion.

5. Initial results for the expected assay precision in random-triggered multiplicity analysis are intriguing, but further analysis and extensive benchmarking is required before any conclusions can be drawn.

6. This initial study of quad coincidences suggests that further investigation into potential applications is warranted. It might be worthwhile to modify other software packages used for multiplicity research and development to collect quad coincidence data. However, it would be premature to incorporate such information into the standard software packages used by DOE facilities.
References


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