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Title: **APPLICATION OF NEUTRON MULTIPLICITY COUNTING TO WASTE ASSAY**

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APPLICATION OF NEUTRON MULTIPLICITY COUNTING TO WASTE ASSAY

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ABSTRACT

This paper describes the use of a new figure of merit code that calculates both bias and precision for coincidence and multiplicity counting, and determines the optimum regions for each in waste assay applications. A "tunable multiplicity" approach is developed that uses a combination of coincidence and multiplicity counting to minimize the total assay error. An example is shown where multiplicity analysis is used to solve for mass, alpha, and multiplication and tunable multiplicity is shown to work well. The approach provides a method for selecting coincidence, multiplicity, or tunable multiplicity counting to give the best assay with the lowest total error over a broad spectrum of assay conditions.

INTRODUCTION

Passive neutron multiplicity counting has been very effective at reducing bias errors in neutron assays. In the hierarchy of neutron counting, the simplest approach is singles counting, which measures neutrons from any source. Next is doubles (or coincidence) counting, which is specific to fission events. The most complex approach is triples (or multiplicity) counting, which is specific to fission and which can also determine multiplication. Neutron multiplicity counting measures singles, doubles, and triples events. From these three measured parameters, it is possible to obtain sample ^{240}Pu mass, the (alpha,n) reaction rate, and sample net leakage multiplication. Or, the three parameters can be used to obtain sample ^{240}Pu mass, (alpha,n) reaction rate, and detector efficiency.

The assay of plutonium metals, oxides, scrap and residues can be significantly improved by using the three measured parameters—single, double, and triple neutron events—to solve for sample mass, (alpha,n) reaction rate, and multiplication. Two recent publications that show good

performance obtained during IAEA inspections of excess weapons materials are for cans of impure oxide[1] and for cans stored in 30-gal. drums.[2]

Application of neutron multiplicity counting to waste assay is suggested because of the significant success of multiplicity counting for safeguards applications. The additional information (compared to conventional coincidence counting) could directly improve assay accuracy by correcting for detector efficiency variations caused by waste drum matrix effects. However, multiplicity counting should only be used if it is more effective in the waste application than coincidence counting. Waste measurements are nominally made under different conditions than typical for safeguards. Waste is typically stored in larger containers, with lower plutonium mass and near unity multiplication. Also, passive neutron coincidence counting assay of waste drums using the segmented add-a-source method has demonstrated the capability for correcting for matrix effects.[3]

In multiplicity counting, singles are measured with the best precision, doubles with moderate precision, and triples with the least precision. Coincidence counting will always have better precision than multiplicity counting because of the additional variance from the triples measurement. On the other hand, waste drum counters have now been designed with high absolute neutron detection efficiencies on the order of 30%.[4,5] With such high detection efficiencies, measurement precision of 1 to 5% can be obtained for triple coincidence events.[6] The basic issue is that for safeguards assay, the small additional error from triples counting is insignificant compared to the improvement in bias error from the multiplicity analysis. For waste applications, the situation may be reversed.

When should coincidence counting be used rather than multiplicity counting? This paper examines this question using a new figure of merit code that calculates both bias and precision for these two techniques, and determines the optimum regions for each. Also, a "tunable multiplicity" approach is described that uses an appropriate combination of coincidence and multiplicity counting to minimize the total assay error. The tunable multiplicity approach is used to predict the total assay error as a function of plutonium mass for typical waste drum applications.

THE POINT MODEL EQUATIONS FOR NEUTRON MULTIPLICITY COUNTING

The point model for neutron multiplicity counting assumes that sample self-multiplication, (alpha,n) reaction rate, detection efficiency, and die-away time are constant across the sample volume, as though the sample were a point. Under these assumptions, the detected singles,

doubles, and triples count rates (with room background removed) are given by the following equations:[7,8,9]

$$S = F\varepsilon M \nu_{s1}(1+\alpha) \tag{1}$$

$$D = \frac{F\varepsilon^2 f_d M^2}{2}\left(\nu_{s2} + \left(\frac{M-1}{\nu_{i1}-1}\right)\nu_{s1}(1+\alpha)\nu_{i2}\right) \tag{2}$$

$$T = \frac{F\varepsilon^3 f_t M^3}{6}\left(\nu_{s3} + \left(\frac{M-1}{\nu_{i1}-1}\right)\left[3\nu_{s2}\nu_{i2} + \nu_{s1}(1+\alpha)\nu_{i3}\right] + 3\left(\frac{M-1}{\nu_{i1}-1}\right)^2 \nu_{s1}(1+\alpha)\nu_{i2}^2\right) \tag{3}$$

where F = sample fission rate,

ε = detector efficiency,

f_d = double gate fraction,

f_t = triples gate fraction,

M = sample net leakage multiplication,

α = ratio of (alpha,n) to spontaneous fission neutrons,

ν_{si} = spontaneous fission moments,

ν_{Ii} = induced fission moments.

Equations (1), (2), and (3) relate the mass, alpha, efficiency, and leakage multiplication to the statistical measure of:

- single neutrons (uncorrelated),
- doubles or coincidences (2 neutrons),
- triples (3 neutrons).

Also note that the statistical differences between the measured count rates are increased by the rate of accidental coincidences.

Equations (1), (2), and (3) are inverted to obtain mass, alpha, and detection efficiency as shown in Eqs. (4), (5), and (6) below. Or, they can be inverted to yield sample mass, alpha, and multiplication as shown in Eqs. (7), (8), and (9) below:

$$\alpha = \frac{3STv_{s2}^2}{2D^2 v_{s1} v_{s3}} - 1 \tag{4}$$

$$F = \frac{2f_d D^3 v_{s3}^2}{9T^2 v_{s2}^3} \tag{5}$$

$$\varepsilon = \frac{3Tv_{s2}}{f_d D v_{s3}} \tag{6}$$

$$F = \frac{\dfrac{2D}{\varepsilon f_d} - \dfrac{M(M-1)v_{i2}S}{v_{i1}-1}}{\varepsilon M^2 v_{s2}} \tag{7}$$

$$\alpha = \frac{S}{F\varepsilon v_{s1} M} - 1 \tag{8}$$

$$a + bM + cM^2 + M^3 = 0 \tag{9}$$

The constants a, b, c in Equation (9) are defined in Ref. 6. Finally, the sample ^{240}Pu effective mass m = F/479.

APPLICATION OF MULTIPLICITY COUNTING TO WASTE

The calculations presented in this paper were obtained using a new figure of merit code developed for multiplicity counting analysis, based on an earlier variance code developed by Ensslin, et. al.[7] The new code described in this paper is able to evaluate both bias and precision for multiplicity and coincidence counting. The code determines assay variance from the reduced factorial moments of the neutron multiplicity distribution, which may be thought of as single, double, and triple neutron coincidences. The multiplicity distribution does not need to be measured, but is predicted from pre-selected sample and detector design parameters. The procedure can be summarized as follows:

1. Input the mass, multiplication, alpha, count time, and detector parameters such as efficiency.

2. Calculate the point model neutron moments: singles, doubles, and triples.
3. Calculate the emitted and detected factorial moments of the neutron distribution function.
4. Calculate the variance in the singles, doubles, and triples.
5. Calculate the sensitivity of the measurement to errors in the singles, doubles, and triples.
6. Calculate the total assay variance from this sensitivity.
7. Calculate the assay mass and the bias in the assay (by using Eqs. 4-9).

One example of the use of the new figure of merit code is given in Fig. 1. This figure compares precision, bias, and total error for multiplicity and coincidence counting for nominal values in safeguards and waste applications. In the safeguards application, the sample mass is assumed to be 1 g ^{240}Pu, with multiplication of 1.2, α of 1, counted for 100 seconds in a neutron counter with 30% detection efficiency. For coincidence counting, assay precision is about 1%, bias is about 280%, and total assay error is about 280%. For multiplicity counting, assay precision is about 5%, bias is close to 0%, and total assay error is 5%. These error estimates assume that it is not feasible to use a non-linear calibration curve to remove the bias in the coincidence count due to (alpha,n) induced fissions, and that the multiplicity assay removes this bias entirely. Therefore, in this case multiplicity has a much lower error, 5%, than coincidence counting, 280%. Multiplicity counting is the better choice in this instance. In the waste application, the sample mass is assumed to be 0.01 g ^{240}Pu, with multiplication of 1.0, α of 1, counted for 100 s in a low-background neutron counter with 30% detection efficiency. For coincidence counting, assay precision is about 15%, bias is nearly 0%, and total assay error is about 15%. For multiplicity counting, assay precision is about 25%, bias is nearly 0%, and total assay error is 25%. In this case, there is no benefit to multiplicity counting; coincidence counting has the lower net error. This figure demonstrates the general issue we wish to address. In some assay regimes (for example, nominal safeguards) multiplicity counting is clearly superior to coincidence counting. In other assay regimes, there is no benefit to multiplicity counting.

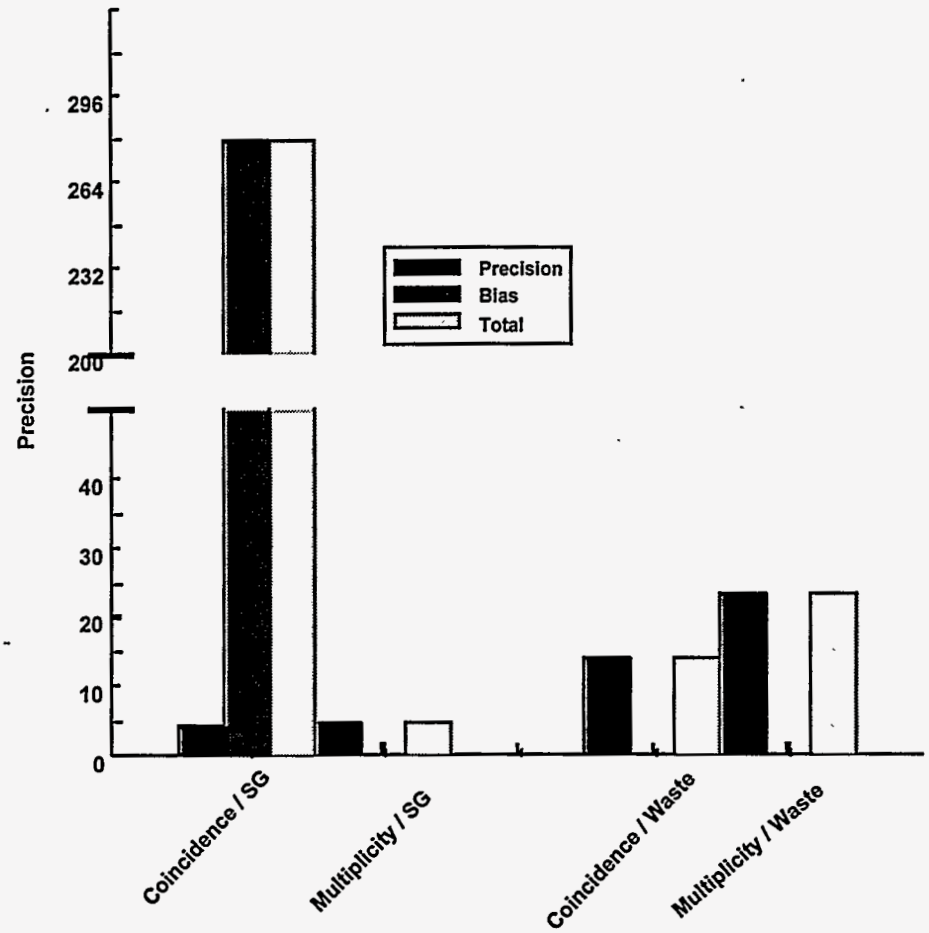

Fig 1. Measurement errors calculated by the figure of merit code for four cases: coincidence and multiplicity counting for a nominal safeguards (SG) measurement of 1 g ^{240}Pu, multiplication of 1.2, alpha of 1, counted for 100 s in a neutron counter with 30% efficiency. The error for these two measurements is dominated by the bias error in the coincidence measurement. The multiplicity case has a slightly larger precision, but the dominant bias error is gone. The second two cases are the same measurement conditions as the first except the mass is 0.01 g ^{240}Pu and the multiplication is 1; this is nominal waste assay conditions. In these cases the bias error is negligible and the dominant error is the precision. The multiplicity measurement has a slightly degraded precision because ofthe addition of the triples measurement.

Figure 2 demonstrates the comparison between multiplicity and coincidence counting again. The figure of merit code is used to determine the total assay error in % by scaling over the range of ^{240}Pu effective from 0.01 to 10.0 g and sample multiplication from 1 to 1.05, with $\alpha = 1$. The sample multiplication is scaled to the sample mass with a quadratic equation. This range of parameters covers most typical waste applications, except for high-alpha wastes such as fluorides. The total error in the coincidence assay is the counting precision and the bias error in quadrature. The total error in the multiplicity assay is just the statistical precision, which is dominated by the poor precision in the triples count rate (the bias is added in quadrature but it is zero). Note that the

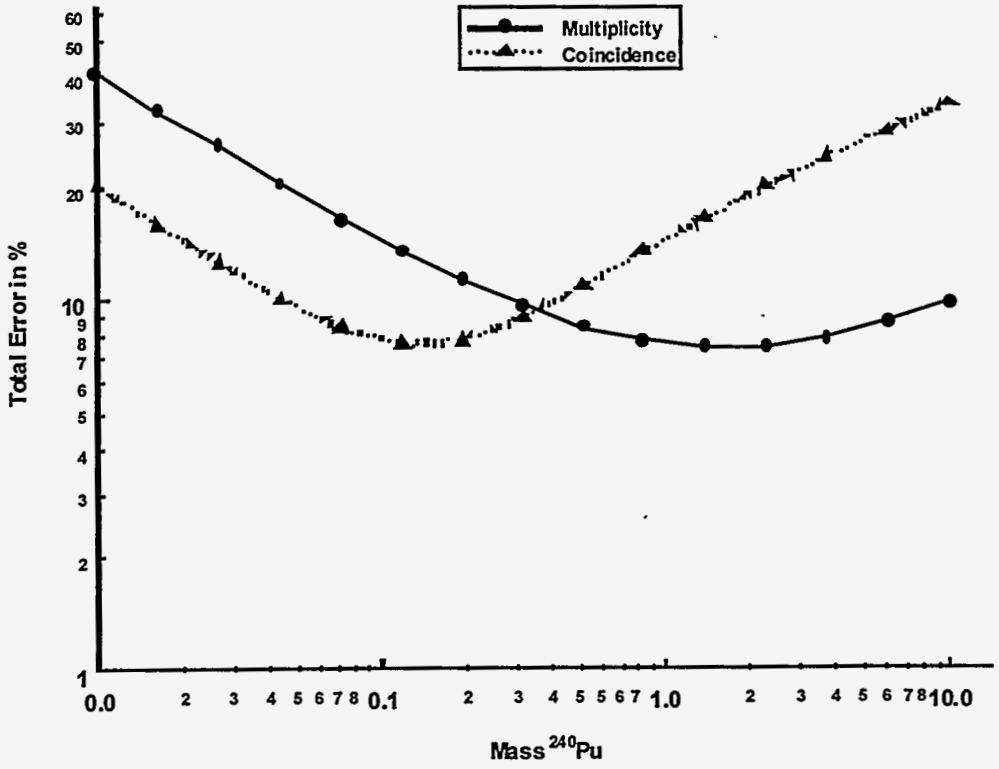

Fig. 2. Application of the new figure of merit code to scale the total error in coincidence and multiplicity counting over the range of ^{240}Pu effective from 0.01 to 10.0 g and sample multiplication from 1 to 1.05 with $\alpha = 1$, for waste assay with 30% detection efficiency.

coincidence assay has a lower total error for low masses with low multiplication, whereas the multiplicity assay has a lower total error for higher masses with higher multiplications.

The individual coincidence components of error, including the bias, precision, and total error are detailed in Fig. 3 for coincidence counting and the same measurement conditions as above. This figure shows how the coincidence bias increases with increasing multiplication, as the induced fission rate increases and biases the coincidence count upwards. The coincidence counting precision improves with increasing mass, eventually leveling off. The bias and precision components combine in quadrature to give a total error with a minimum near 0.10 g ^{240}Pu effective.

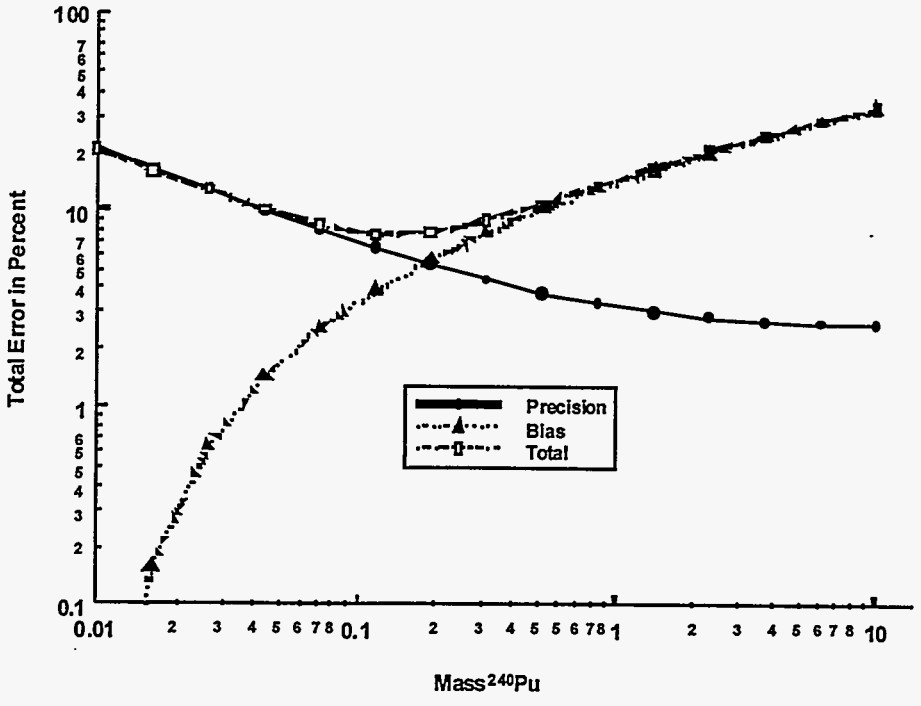

Fig. 3. The coincidence bias, precision, and total error over the range of ^{240}Pu effective from 0.01 to 10.0 g and sample multiplication from 1 to 1.05 with $\alpha = 1$, for waste assay with 30% detection efficiency.

By way of comparison, Fig. 4 illustrates the error sources that make up the total multiplicity error. Only counting statistics error sources are shown, although multiplicity counting can also exhibit bias for actual samples if the assumptions of the point model are not valid. Figure 4 shows the contribution to the variance from singles, doubles, and triples for safeguards applications with ^{240}Pu mass of 1 g, multiplication of 1.2, and alpha of 1 for a neutron counter with 20% detection efficiency; also for waste applications with multiplication = 1.0. The triples counting precision dominates the total error, as expected from the significantly lower triples count rate. Note that the curve of total error for multiplicity counting shown earlier in Fig. 2 also has a minimum, although it is caused only by counting precision, because the accidental overlap rate of multiplicity events increases with increasing sample mass.

COMPARISON OF CALCULATED AND OBSERVED PRECISION

To benchmark the figure of merit calculations described above, we compared observed and calculated assay precision for a series of small plutonium samples. Each sample contained 10 g

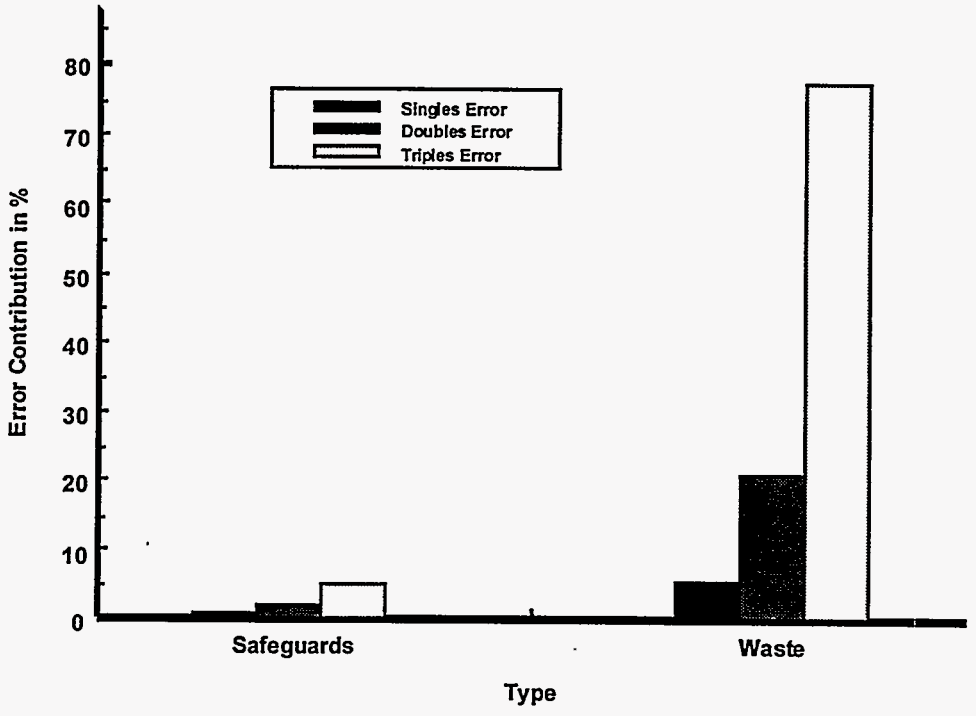

Fig. 4. Multiplicity error sources from singles, doubles, and triples counts for nominal safeguards and waste assay applications, with 20% detection efficiency.

of plutonium, but the plutonium was intimately mixed with an impurity element that was a source of (alpha,n) neutrons. The impurity elements were pure metal (no impurities), oxide, aluminum, silicon, magnesium, and fluorine. These sample matrices simulate the range of neutron-emitting impurities that can be found in actual transuranic waste drums. Observed assay precision was obtained by placing the samples in the center of empty 55-gal. waste drums and carrying out a series of passive neutron coincidence and multiplicity measurements using the waste drum counters described in Ref. 4 and 5. Each sample was measured for a long series of short runs, typically 30 to 100 runs of 100 s each.

Observed variances (normalized to 1000-s counting times) ranged from 1 to 16% for coincidence analysis, from 2 to 22% for multiplicity analysis solving for sample multiplication, and from 11% to 39% for multiplicity analysis solving for detection efficiency. The observed RSD was larger than the calculated RSD by about 30% for conventional coincidence counting and 35% for multiplicity counting. These results are consistent with past benchmarking studies, and are due to the fact that the effects of correlations between detected neutron events are not included in the computation of variance in the calculated multiplicity distributions. When these correlations are

taken into account, we can expect the figure of merit calculations in this paper to be accurate to about 10 to 15%, which is sufficient for observing the effects of parameter variations.

NEW INVERSION OF THE MULTIPLICITY EQUATIONS

A comparison of the error structure in Fig. 2 shows that multiplicity is better when the bias is high and the precision is good. On the other hand, coincidence is better when the bias is low, as is the case in many waste applications. Figure 2 suggests the potential benefits of a new approach that could minimize assay error by selecting either coincidence or multiplicity counting, as appropriate. Is there a heuristic system that can determine from the data when to use coincidence and when to use multiplicity?

Equations (4) through (9) in Section II show a discrete change in the mathematics from coincidence to multiplicity. There is no gradual shift from one to the other, the choice is either coincidence or multiplicity. We also observe that the multiplicity equations are inverted algebraically. In the inversion, no distinction is made between singles, doubles and triples. All three parameters are used equally. The inversion does not weight the relative errors in the singles, doubles, or triples, even through the triples variance is typically higher, as was shown in Fig. 4.

These observations suggest another approach: a perturbation expansion inversion of the point equations. The singles, doubles, and triples could be weighted according to their variance. In this way we could include "partial" multiplicity, and we could move continuously between coincidence and multiplicity. This approach will be called "tunable multiplicity." The tunable multiplicity approach will use a perturbation expansion of the equations, and then linearize them by keeping only first order terms. The perturbation can be adjusted with a multiplicative constant to move from zero perturbation (coincidence counting) to full perturbation (multiplicity counting to first order). Two cases will be solved, inverting the multiplicity equations for mass, α, and efficiency and inverting for mass, α, and multiplication.

To solve for efficiency, the efficiency is expanded about ε_0, the unperturbed detection efficiency of the waste drum counter with no sample matrix present. The triples to doubles ratio T/D is used as the expansion parameter and only first order terms are kept. The resulting inverted equations are

$$\varepsilon\left(S,D,T/D\right) \cong \varepsilon_o + \frac{\partial \varepsilon}{\partial\left(\frac{T}{D}\right)}\left[\left(\frac{T}{D}\right) - \left(\frac{T}{D}\right)\Big|_{\varepsilon=\varepsilon_o}\right] \tag{10}$$

Use of Eq. (10) results in the following new inverted equations:

$$\alpha = \frac{3STv_{s2}^2}{2D^2 v_{s1} v_{s3}} - 1 \tag{11}$$

$$F = \frac{2D}{f_d v_{s2} \varepsilon_o^2} - \frac{4D}{f_d v_{s2} \varepsilon_o^3}\left(\frac{3v_{s2}T}{f_d v_{s3} D} - \varepsilon_o\right) \tag{12}$$

$$\varepsilon_1 = \frac{3v_{s2}T}{f_d v_{s3} D} - \varepsilon_o \tag{13}$$

To solve for multiplication the equation for multiplication will be expanded about $M = 1$, again using T/D as the expansion parameter:

$$M\left(S,D,T/D\right) = M_o + \frac{\partial M}{\partial\left(T/D\right)}\left[\left(\frac{T}{D}\right) - \left(\frac{T}{D}\right)\Big|_{M\equiv 1}\right] \tag{14}$$

Use of Equation (14) results in the following new inverted equations:

$$F = \frac{2D}{\varepsilon_o^2 f_d v_{s2}} - \frac{M_1}{\varepsilon_o v_{s2}}\left[\frac{4D}{\varepsilon_o f_d} + \frac{S v_{i2}}{(v_{i1} - 1)}\right]\beta \tag{15}$$

$$\alpha = \frac{S}{F\varepsilon_o v_{s1}(1 + M_1\beta)} - 1 \tag{16}$$

$$M = 1 + M_1\beta \tag{17}$$

$$M_1 = \frac{2D(v_{i1} - 1)\left[\varepsilon_o f_d v_{s3} - 3v_{s2}\left(T/D\right)\right]}{\varepsilon_o f_d\left[3Sv_{s2}v_{i2}(T/D) - 2D(v_{i1} - 1)v_{s3} - 6Dv_{s2}v_{i2} - Sv_{i3}v_{s2}\varepsilon_o f_d\right]} \tag{18}$$

The parameter β in equations (15), (16), and (17) represents the ability to tune the amount of the multiplicity contribution. A β factor of 0 is zero perturbation (coincidence only) and a beta

factor of 1 is full perturbation (multiplicity counting to first order). The optimization of the tunable multiplicity is illustrated in Fig. 5, which plots precision, bias, and beta as a function of ^{240}Pu effective mass. The figure of merit code is used to scale the mass and multiplication and calculate the total error in % over the range of ^{240}Pu effective from 0.01 to 10.0 g and sample multiplication from 1 to 1.05, with $\alpha = 1$, detection efficiency = 20%, and counting time = 100 s. The sample multiplication is again scaled to the sample mass with a quadratic equation. As the figure of merit code steps through the values of sample mass and multiplication, the beta factor is also varied to find the value that gives the lowest total error. Note that at very low mass, where coincidence counting has a lower total error (ref. Fig. 2), the beta factor is only about 10 or 15%, which is nominaly consistent with coincidence counting. At high mass, where multiplicity counting has a lower total error, the beta factor is 70 or 80%, so that more multiplicity information is being tuned in to reduce the total error.

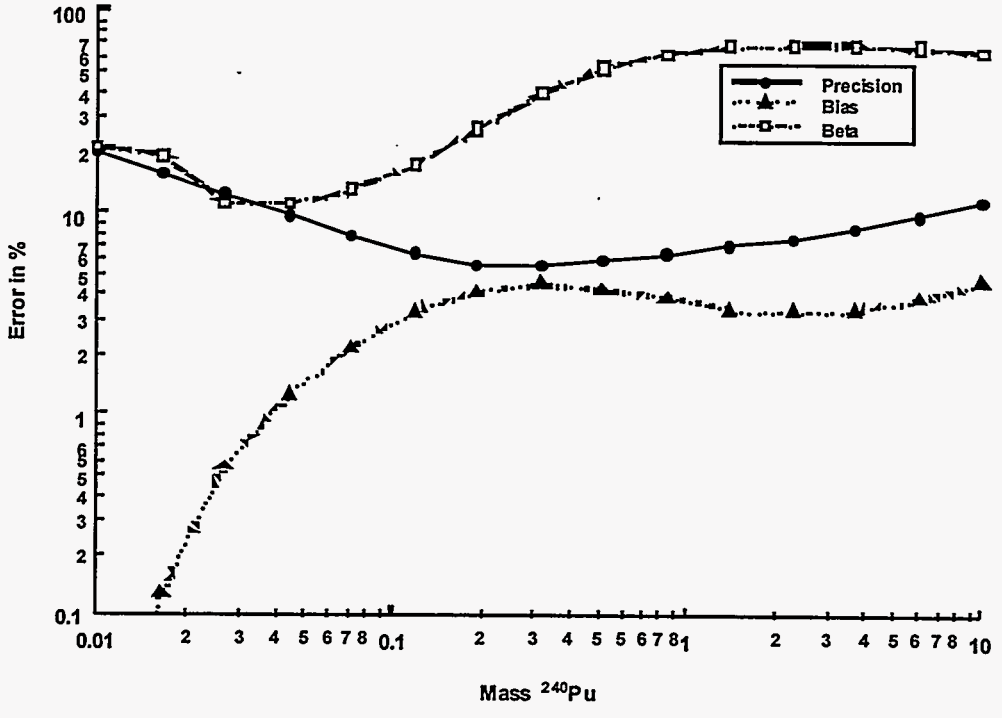

Fig. 5. Optimization of the tunable multiplicity using the new figure of merit code. The percent error in precision and bias, and the beta factor in percent, are scaled over the range of ^{240}Pu effective from 0.01 to 10.0 g and sample multiplication from 1 to 1.05, with $\alpha = 1$, detection efficiency = 20%, and counting time = 100 s.

Figure 6 compares the total error calculated from the figure of merit code for three cases: coincidence, multiplicity, and tunable multiplicity for the same measurement parameters (mass, multiplication, α, etc.) as Fig. 5. Note that as the sample mass and multiplication vary over the

allowed range, the tunable multiplicity approach yields the lowest total error for most mass values, and is close to the lowest for the others.

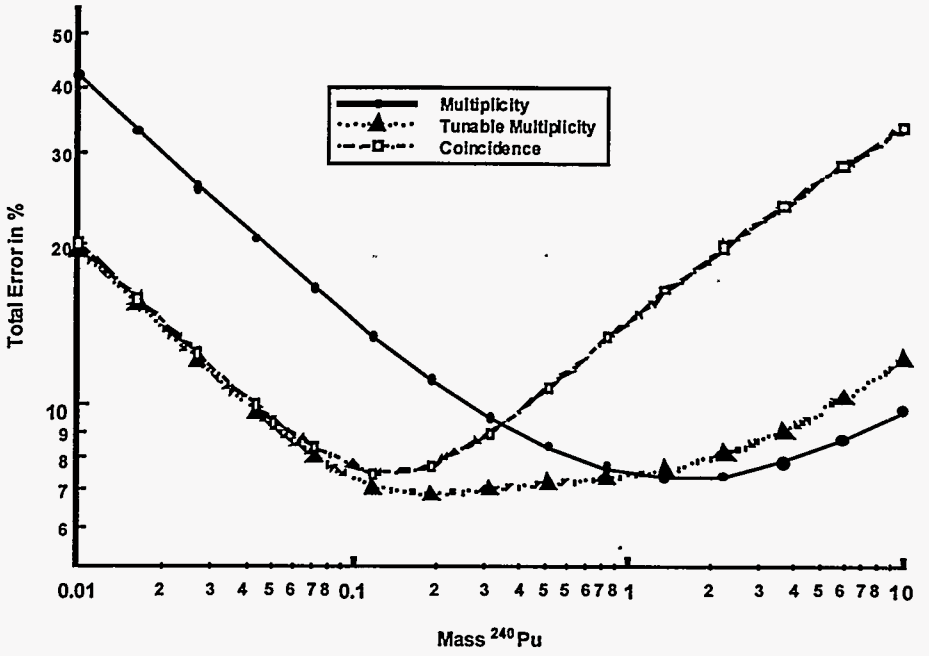

Fig. 6. Comparison of the total error in tunable multiplicity with coincidence and multiplicity counting using the figure of merit code, for the same range of measurement parameters as Fig. 5.

CONCLUSIONS

1. It has been possible to develop a tunable multiplicity approach for combining coincidence and multiplicity analysis for waste drum assay. A new figure of merit code has also been developed to evaluate the tunable multiplicity approach.

2. For the case where multiplicity analysis is used to solve for mass, alpha, and multiplication, the tunable multiplicity approach works well. We are able to move continuously between coincidence counting and multiplicity, and obtain the optimal lowest total error in most cases. The cusp observed in Figs. 2 and 6 at the transition from coincidence to multiplicity is smoothed out, and a total error less than the cusp value is obtained. As assay bias increases, the multiplicity "turns on," as expected.

3. A method for choosing the beta factor from actual measurement data in a field environment needs to be developed.

4. The tunable multiplicity approach described above needs to be extended to the case where multiplicity analysis is used to solve for detection efficiency.

5. The approach described in this paper leads directly to a heuristic method for selecting coincidence, multiplicity, or tunable multiplicity counting as the best assay approach with the lowest total error.

ACKNOWLEDGMENTS

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