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EXPECTED PRECISION OF NEUTRON MULTIPLICITY MEASUREMENTS OF WASTE DRUMS[*]

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ABSTRACT

DOE facilities are beginning to apply passive neutron multiplicity counting techniques to the assay of plutonium scrap and residues. There is also considerable interest in of applying this new measurement technique to 208-liter waste drums. The additional information available from multiplicity counting could flag the presence of shielding materials or improve assay accuracy by correcting for matrix effects such as (α,n) induced fission or detector efficiency variations. The potential for multiplicity analysis of waste drums, and the importance of better detector design, can be estimated by calculating the expected assay precision using a Figure of Merit code for assay variance. This paper reports results obtained as a function of waste drum content and detector characteristics. We find that multiplicity analysis of waste drums is feasible if a high-efficiency neutron counter is used. However, results are significantly poorer if the multiplicity analysis must be used to solve for detection efficiency.

I.   INTRODUCTION

Passive neutron multiplicity counting can significantly improve the assay of plutonium metals, oxides, scrap, and residues by using three measured parameters—single, double, and triple neutron events—to solve for sample mass, self-multiplication, and (α,n) reaction rate (Ref. 1). However, the triples neutron count rate is proportional to the third power of the neutron detection efficiency, so that a high-efficiency neutron counter is needed to obtain a statistically precise value for the observed triples count rate. Most of the neutron multiplicity counters built for the assay of plutonium cans have absolute neutron detection efficiencies in the range of 40 to 55%.

By contrast, most neutron counters built for the assay of plutonium waste in 55-gal. drums have detection efficiencies in the range of 15 to 18%. Also, waste drums may contain only a few grams of plutonium, rather than bulk quantities. Both of these

considerations suggest that the triples count rate in waste might be too low to obtain good safeguards assays in reasonable counting times. However, the potential benefits of multiplicity analysis of waste are intriguing. The additional information available from multiplicity counting could be used to monitor the neutron detection efficiency, thereby flagging the presence of shielding materials. Or, the neutron triples/doubles ratio could detect the presence of highly multiplying samples that should not be present in waste. Also, the additional information could directly improve assay accuracy by correcting for matrix effects such as (α,n) induced fission or detector efficiency variations.

New waste drum counters with substantially higher neutron detection efficiencies are now under development (Ref. 2), so that multiplicity assay of waste may become possible with good throughput rates. For example, Canberra Industries and Los Alamos National Laboratory are carrying out a Cooperative Research and Development Agreement (CRADA) to build an Add-a-Source Waste Drum Assay System with a neutron detection efficiency on the order of 35%. Because of the potential benefits of multiplicity analysis for safeguards accountability and diversion detection, and the importance of taking this into account during the detector design process, this paper presents a series of calculations that estimate the expected assay precision for multiplicity assay of waste. We report results obtained as a function of waste drum content and detector characteristics for two modes of analysis: using the extra multiplicity information to determine sample self-multiplication, or using the information to obtain the neutron detection efficiency of the counter with the waste drum inside. We compare our results with some available laboratory measurements and provide some conclusions on the applicability of this technique to waste assay.

II.   PROCEDURE FOR CALCULATING MULTIPLICITY ASSAY PRECISION

The calculations presented in this paper were obtained using a Figure of Merit code developed for multiplicity counting analysis (Ref. 3). 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

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neutron coincidences. The multiplicity distribution does not need to be measured but is predicted from pre-selected sample and detector design parameters. In the past, the Figure of Merit code has only been applied to determine assay variance for samples with unknown mass, self-multiplication, and (α,n) reaction rate, where all detector parameters including detection efficiency are assumed to be known. In this situation, the procedure can be summarized as follows (Refs. 3,4):

1. The detected singles, doubles, and triples count rates (with room background removed) are given by

$$S = mF\varepsilon M v_{s1}(1+\alpha)$$

$$D = \frac{mF\varepsilon^2 f_d M^2}{2}\left[v_{s2} + \left(\frac{M-1}{v_{i1}-1}\right)v_{s1}(1+\alpha)v_{i2}\right]$$

$$T = \frac{mF\varepsilon^3 f_t M^3}{3} \cdot$$

$$\left\{v_{s3} + \left(\frac{M-1}{v_{i1}-1}\right)[3v_{s2}v_{i2} + v_{s1}(1+\alpha)v_{i3}]\right.$$

$$\left. + 3\left(\frac{M-1}{v_{i1}-1}\right)^2 v_{s1}(1+\alpha)v_{i2}{}^2\right\} ,$$

where

m = effective ^{240}Pu mass,

F = ^{240}Pu spontaneous fission rate,

ε = neutron detection efficiency,

M = neutron multiplication,

α = (α,n) to spontaneous fission neutron ratio

f_d = doubles gate fraction,

f_t = triples gate fraction,

v_{s1}, v_{s2}, v_{s3} = first, second, and third reduced moments ofspontaneous fission neutron distribution,

v_{i1}, v_{i2}, v_{i3} = first, second, and third reduced moments of the induced fission neutron distribution, and

S, D, T = singles, doubles, and triples rates.

Equations (1), (2), and (3) are solved for m, M, and α as follows:

$$a + bM + cM^2 + M^3 = 0 ,$$

where

$$a = \frac{-3Tv_{s2}(v_{i1}-1)}{\varepsilon^2 f_t S(v_{s2}v_{i3} - v_{s3}v_{i2})} , \qquad (4)$$

$$b = \frac{2D[v_{s3}(v_{i1}-1) - 3v_{s2}v_{i2}]}{\varepsilon f_d S(v_{s2}v_{i3} - v_{s3}v_{i2})} ,$$

and

$$c = \frac{6Dv_{s2}v_{i2}}{\varepsilon f_d S(v_{s2}v_{i3} - v_{s3}v_{i2})} - 1 .$$

$$F = \frac{\left[\dfrac{2D}{\varepsilon f_d} - \dfrac{M(M-1)v_{i2}S}{v_{i1}-1}\right]}{\varepsilon M^2 v_{s2}} \qquad (5)$$

$$m = F/479 \qquad (6)$$

$$\alpha = \frac{S}{F\varepsilon v_{s1}M} - 1 . \qquad (7)$$

Finally, the assay variance is given by

$$dF^2 = \left(\frac{\partial F}{\partial S}\right)^2 dS^2 + \left(\frac{\partial F}{\partial D}\right)^2 dD^2 + \left(\frac{\partial F}{\partial T}\right)^2 dT^2 + 2\left(\frac{\partial F}{\partial D}\right)\left(\frac{\partial F}{\partial T}\right)\text{COV}^2.$$ (8)

The relative standard deviation (RSD) in % is given by

$$\text{RSD } (\%) \text{ in } F = 100 * dF/F.$$ (9)

In Eq. (8), the errors dS, dD, and dT and the covariance term COV are determined by detailed computations of the estimated factorial moments of the expected multiplicity distributions (Ref. 3). On the other hand, the partial derivatives in Eq. 8 are determined simply by varying S, D, and T by small arbitrary amounts and calculating the resulting change in F, M, and α using equations (4) through (7).

The above procedure was used in Section IV below to determine assay precision when multiplicity analysis is used to determine mass, multiplication, and the (α,n) reaction rate α from S, D, and T, with detector efficiency assumed known. However, for Section V below we need to determine mass, detector efficiency, and α from S, D, and T with multiplication M assumed to be 1. For this situation we replace Eqs. (4) through (7) by the following equations:

$$\alpha = \frac{3\,STv_{s2}^2}{4\,D^2 v_{s1} v_{s3}} - 1$$ (10)

$$F = \frac{S^2 f_d v_{s2}}{D v_{s1}^2 (1+\alpha)^2}$$ (11)

$$m = F / 479$$ (12)

$$\varepsilon = \frac{S}{F v_{s1}(1+\alpha)}$$ (13)

The variance estimation procedure is the same, except that when we vary S, D, and T, we calculate the resulting change in F, ε, and α using Eqs. (10) through (13).

III. COMPARISON OF CALCULATED AND OBSERVED PRECISION

To benchmark the current versions of the Figure of Merit code against data taken recently with multiplicity counters, we compared calculated and observed assay precisions for a variety of plutonium metal and oxide samples. The samples ranged in mass from 0.53 to 142 g of Pu-240 effective, in multiplication from 1.0076 to 2.044, and in α from 0 to 4.88. Because the current version of the Figure of Merit code does not accept the actual, experimentally determined gate fractions f_d and f_t, the input values for the die-away time were adjusted by small amounts so that the code would be working with singles, doubles, and triples count rates close to the measured ones.

If multiplicity analysis is used to determine mass, M, and α, we compared the Figure of Merit code with data from a Plutonium Scrap Multiplicity Counter (PSMC). The PSMC had an efficiency of 55%, a gatewidth of 32 μs, and a die-away time of 47 μs. If multiplicity analysis is used to determine mass, detector efficiency, and α, we compared the Figure of Merit code with data from a three-ring Active Well Coincidence Counter adapted for multiplicity analysis. The three-ring counter had an efficiency of 50%, a gatewidth of 32 μs, and a die-away time of 75 μs.

In both cases, we found that the observed RSD is higher than the calculated RSD by about 10 to 15% for most samples. For very highly multiplying samples, the observed RSD is higher by about 50%. Both results are consistent with the fact that the computation of variance from the calculated multiplicity distributions does not include the effect of correlations between events. In the case of multiplicity assay of waste, where there are relatively few correlated events, we can expect the code to yield results good to within 10 to 15%, which is sufficient for predicting assay precision, count time, and optimum design parameters.

IV. CALCULATED PRECISION WHEN MULTIPLICITY IS USED TO SOLVE FOR MASS, MULTIPLICATION, AND ALPHA

The neutron multiplicity hardware, software, electronics, and data analysis algorithms developed at Los Alamos are intended to provide DOE facilities with a means of quickly assaying impure plutonium samples such as oxidized metals, dirty oxides, and scrap and residue materials. The third measured parameter is needed to correct for the variable (α,n) reaction rates in these impure materials. The cans containing the impure plutonium are not very large, and the matrix materials in the cans do not appreciably

perturb the counter's efficiency for detecting emitted neutrons. Also, the ratios of total neutron counts in the different ^{3}He detector rings can be used to make small corrections to the efficiency (Ref. 5). Thus the detector efficiency can be considered a known parameter, and the measured singles, doubles, and triples count rates can be used to determine sample mass, M, and α. Past examples of the expected counting precision for this situation are given in Refs. 3 and 6.

For the multiplicity assay of waste samples using this approach, Eqs. (1) through (9) in Section II are used to calculate the expected assay precision. The sample mass is anticipated to be in the range of 1 to 100 g of plutonium, or roughly 0.1 to 10 g of ^{240}Pu. A new high-efficiency waste drum counter is expected to have an absolute neutron detection efficiency in the range of 25 to 35% or more, with a die-away time in the range of 70 to 120 μs. The expected assay precision in this range of performance is illustrated in Figs. 1 through 4 for several important sample and detector parameters. Each figure plots the expected assay RSD (%) as a function of ^{240}Pu mass for 1000-s counting times. In all cases, the shift register electronics predelay is assumed to be 3 μs, and the detector electronics deadtime is assumed to be 100 ns. Sample self-multiplication is set to 1, which is conservative in the sense that a slightly higher value for M would yield more triple coincidences and thereby lower the assay RSD.

Figure 1 illustrates neutron totals, conventional coincidence, and multiplicity RSD for two values of counter efficiency, 25% and 35%. This figure is a "best case" in the sense that optimum low values have been selected for sample α, counter background, and counter die-away time. The comparison of conventional coincidence and multiplicity RSD illustrates the "penalty" in RSD that results from utilizing multiplicity analysis. Note that coincidence RSD levels off with increasing mass, whereas multiplicity RSD passes through a minimum and then rises with increasing accidental coincidence background. In Fig. 1, however, multiplicity analysis still yields good RSD values of 1 to 4% over the expected mass range, for both values of detector efficiency.

Figure 2 illustrates neutron coincidence and multiplicity RSD for three values of sample α, with 35% detector efficiency and a moderate die-away time of 100 μs. The figure shows that multiplicity assay of relatively clean plutonium waste, with α values between 0.5 and 5, will have a RSD in the range of 2 to 10%. Waste with high-fluoride content may have α values up to about 20, and the RSD will rise to 20 to 100%. Conventional coincidence counting will have a much lower RSD, but can be biased high by factors of 20% to 500% over this range of sample impurity. Thus the multiplicity assay may prove to

be much more accurate, even though it will be less precise!

Figure 3 illustrates the effect of chamber die-away time and electronics gate width on neutron multiplicity RSD for two values of sample α, with 35% detector efficiency. This complicated graph shows several subtle effects of these two parameters, but the overall picture is that these parameters affect RSD by only a factor of two or less if they are within a reasonable range. In general, a lower die-away time is better, and the RSD is minimized for a gate width that is 1.27 times the die-away time (if the room background is small). However, changing sample α from 1 to 5 will have a larger effect than the effect of chamber die-away time.

Figure 4 illustrates the effect of room background on coincidence and multiplicity RSD. The chamber die-away time is set to 120 μs, the electronics gate width is set to 128 μs, and the detector efficiency is 35%. These large values will be quite sensitive to room background and perhaps represent a "worst case." The figure shows the importance of keeping the room background in the range of 10 to 100 counts/s for either coincidence or multiplicity counting.
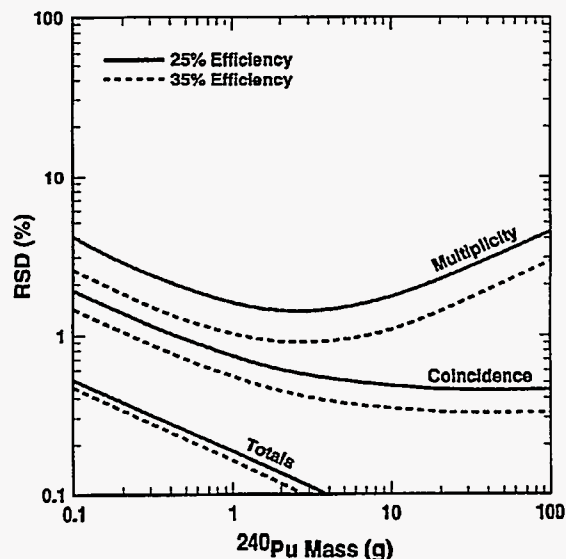

Fig. 1. Assay precision for passive neutron totals, coincidence and multiplicity assay of waste, for the case where the multiplicity information is used to solve for mass, multiplication, and (α,n) reaction rate. Gate width = 64 μs, die-away time = 70 μs, $\alpha = 0.5$, and background = 10 counts/s. This is a "best case" situation.
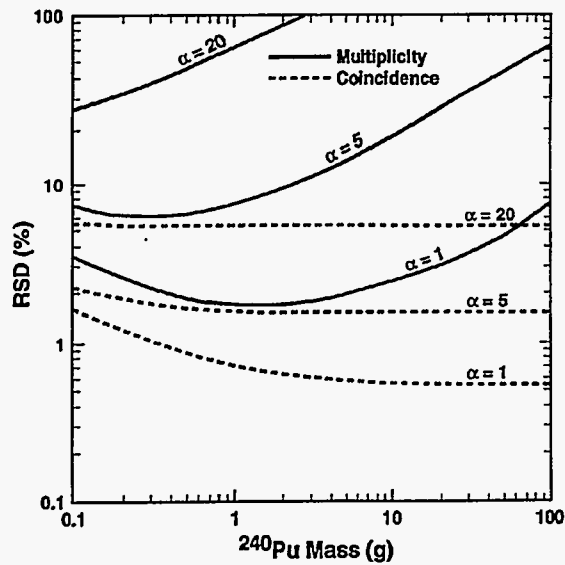
Fig. 2. Assay precision for passive neutron coincidence and multiplicity assay of waste, for the case where the multiplicity information is used to solve for mass, multiplication, and (α,n) reaction rate. Gate width = 64 μs, die-away time = 100 μs, detector efficiency = 35%, and background = 10 counts/s. The dependence of assay precision on α is illustrated.
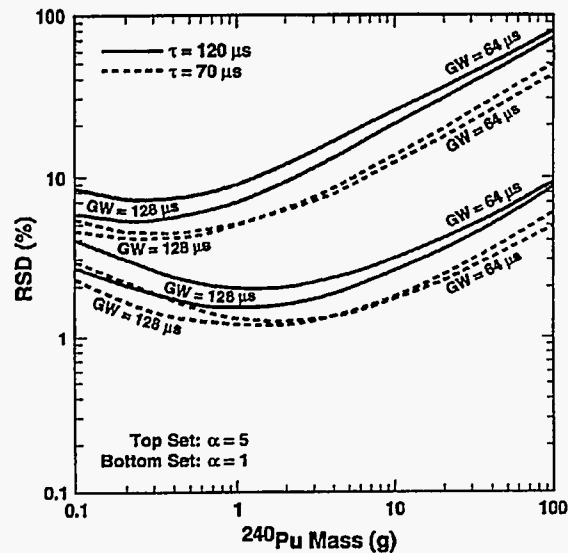

Fig. 3. Assay precision for passive neutron multiplicity assay of waste, for the case where the multiplicity information is used to solve for mass, multiplication, and (α,n) reaction rate. Detector efficiency = 35% and background = 10 counts/s. The dependence of assay precision on gate width and die-away time is illustrated for α = 1 and α = 5.
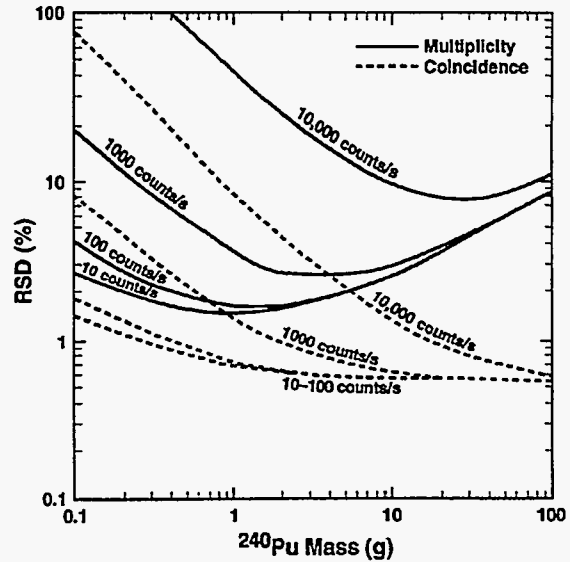

Fig. 4. Assay precision for passive neutron coincidence and multiplicity assay of waste, for the case where the multiplicity information is used to solve for mass, multiplication, and (α,n) reaction rate. Detector efficiency = 35%, gate width = 128 μs, die-away time = 120 μs, and α = 1. The dependence of assay precision on background is illustrated.

V. CALCULATED PRECISION WHEN MULTIPLICITY IS USED TO SOLVE FOR MASS, DETECTION EFFICIENCY, AND ALPHA

At the Joint Research Centre, Ispra, Italy, Cifarelli and Hage have developed multiplicity data analysis algorithms to determine other combinations of three sample parameters (Ref. 7). In particular, the Joint Research Centre has been interested in the assay of large waste drums with relatively low loadings of plutonium. In this situation, it is a good approximation to assume that sample self-multiplication equals 1. However, matrix materials in the large waste containers can significantly affect the counter's neutron detection efficiency. Thus sample self-multiplication can be considered a known parameter, and the measured singles, doubles, and triples count rates can be used to determine sample mass, detection efficiency, and α. For this situation, the expected assay precisions determined from the Figure of Merit code using Eqs. 10-13 are illustrated in Figs. 5 through 8. Each figure employs the same parameters as the corresponding figure in Section IV; sample self-multiplication is again set to 1.
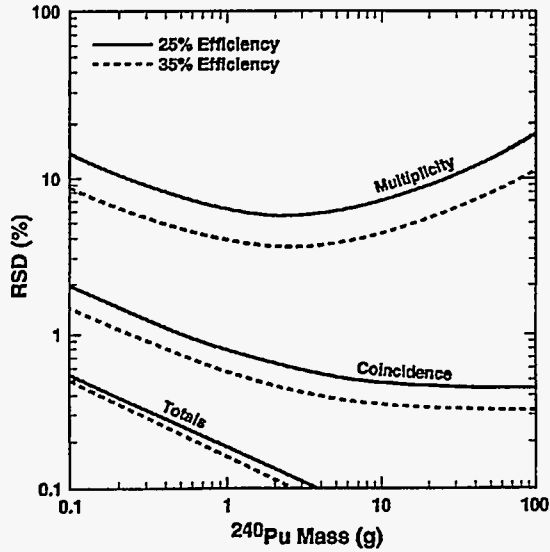
Fig. 5. Assay precision for passive neutron totals, co-incidence, and multiplicity assay of waste, for the case where the multiplicity information is used to solve for mass, detector efficiency, and (α,n) reaction rate. Gate width = 64 μs, die-away time = 70 μs, α = 0.5, and background = 10 counts/s. This is a "best case" situation.
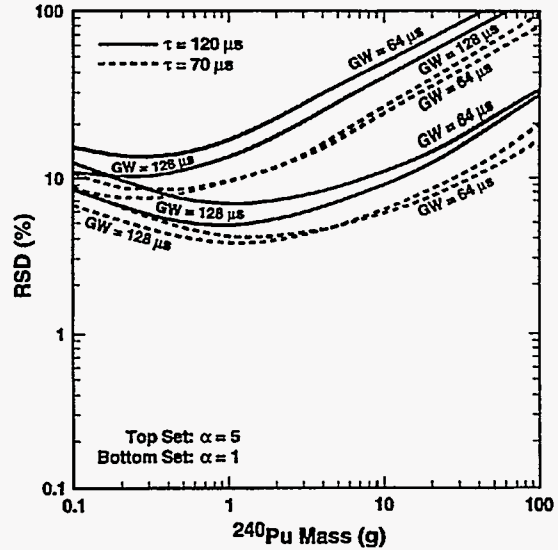

Fig. 7. Assay precision for passive neutron multiplicity assay of waste, for the case where the multiplicity information is used to solve for mass, detector efficiency, and (α,n) reaction rate. Detector efficiency = 35% and background = 10 counts/s. The dependence of assay precision on gate width and die-away time is illustrated for α = 1 and α = 5.

Fig. 6. Assay precision for passive neutron coincidence and multiplicity assay of waste, for the case where the multiplicity information is used to solve for mass, detector efficiency, and (α,n) reaction rate. Gate width = 64 μs, die-away time = 100 μs, detector efficiency = 35%, and background = 10 counts/s. The dependence of assay precision on α is illustrated.
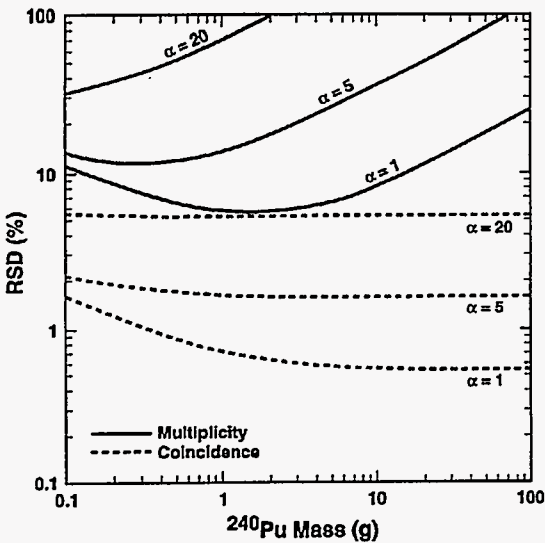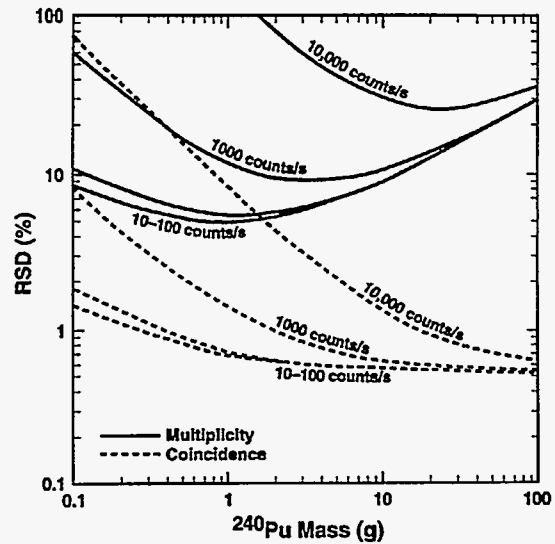

Fig. 8. Assay precision for passive neutron coincidence and multiplicity assay of waste, for the case where the multiplicity information is used to solve for mass, detector efficiency, and (α,n) reaction rate. Detector efficiency = 35%, gate width = 128 μs, die-away time = 120 μs, and α = 1. The dependence of assay precision on background is illustrated.

Figure 5 illustrates neutron totals, conventional coincidence, and multiplicity RSD for two values of counter efficiency, 25% and 35%. This figure should be compared to Fig. 1, and is again a best case in the sense that optimum low values have been selected for sample α, counter background, and counter die-away time. The totals and coincidence RSDs are the same as those in Fig. 1, but the multiplicity RSD is worse by a factor of 3 to 4 over the entire mass range. This surprising result was not expected. However, if Eq. 10 were substituted into Eq. 11, we would see that—when detector efficiency is treated as an unknown—the fission rate F depends on the third power of the doubles rate and the second power of the triples rate. This strong dependence apparently leads to substantially larger partial derivatives in Eq. 8 than are obtained when M is treated as an unknown.

Figure 6 illustrates neutron coincidence and multiplicity RSD for three values of sample α, with 35% detector efficiency and a moderate die-away time of 100 μs. This figure should be compared with Fig. 2, and shows that the RSD is worse by a factor of 3 for $\alpha = 1$, worse by a factor of 2 for $\alpha = 5$, and about the same for $\alpha = 20$. Apparently the loss of precision caused by solving for detector efficiency rather than sample multiplication decreases with increasing α.

Figure 7 illustrates the effect of chamber die-away time and electronics gate width on neutron multiplicity RSD for two values of sample α, with 35% detector efficiency. Figure 7 should be compared with Fig. 3, and again shows that the loss of precision caused by solving for detector efficiency rather than sample multiplication decreases with increasing α.

Figure 8 illustrates the effect of room background on coincidence and multiplicity RSD. The chamber die-away time is set to 120 μs, the electronics gate width is set to 128 μs, and the detector efficiency is 35%. These large values will be quite sensitive to room background and perhaps represent a "worst case." Figure 8 should be compared with Fig. 4. Again it shows the importance of low room background and the loss of precision caused by solving for detector efficiency.

VI.   CONCLUSIONS

1. We have evaluated the potential application of neutron multiplicity analysis to waste drum assay using a Figure of Merit code for assay variance. We estimate that multiplicity assay of relatively clean plutonium waste, with α values between 0.5 and 5, will have a RSD in the range of 2 to 10%. This is not the same high precision and accuracy that multiplicity counters can achieve for the assay of small cans of plutonium, but does show that multiplicity of

waste is feasible if high-efficiency drum counters are used.

2. For waste with very high-impurity content, the multiplicity RSD will rise to 20 to 100%. Even with this poor precision, multiplicity assay may be more accurate than conventional coincidence counting because the bias caused by (α,n) induced fissions is corrected.

3. When we use multiplicity analysis to solve for detector efficiency rather than sample multiplication, we can expect the multiplicity RSD to be increased by a factor of 3 to 4 over the entire mass range. The assay RSD will be 5 to 15% at best. However, the loss of precision caused by solving for detector efficiency decreases with increasing α. For very impure plutonium wastes, both approaches have comparably high RSDs.

4. For multiplicity analysis of waste, it is important to use a high-efficiency waste drum counter. However, it is not crucial that the efficiency be extremely high; an increase in efficency from 25% to 35% reduces the assay RSD by only a factor of two. The drum counter should also have low chamber die-away time and optimum coincidence gate width. These two features will also reduce assay RSD, but only by a factor of two or less.

5. The waste drum counter should be operated with a room background in the range of 10 to 100 counts/s. Higher values will seriously degrade both coincidence and multiplicity assay RSD over the mass range of 1 to 50 g of plutonium.

VII. RECOMMENDATIONS

1. Because the assay RSD for multiplicity of waste is predicted to be reasonably good for many potential samples, we recommend that the technique be thoroughly evaluated for waste drums. It is important to ascertain the range of impure waste drums over which the gain in assay accuracy outweighs the loss of assay precision relative to conventional coincidence counting.

2. Because the use of multiplicity analysis to solve for detector efficiency significantly increases the multiplicity RSD, we should avoid this approach. Where possible, other techniques for determining detection efficiency, such as Segmented Add-a-Source (Ref. 8) or ring-ratio analysis (Ref. 5) should be employed.

3. Because the Figure of Merit code suggests that there is a range of impure plutonium waste with very high α where the multiplicity assay RSD will be no better than 20% or 30%, whether we are solving for multiplication or for detection efficiency, we should evaluate the latter approach for this case. Solving for detector efficiency may provide a direct correction for neutron energy spectrum shifts caused by the neutrons from (α,n) reactions.

ACKNOWLEDGMENTS

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