Comparison Of Neutron Coincidence And Multiplicity Counting Techniques For Safeguards

Dorothy Davidson and Robert McElroy
CANBERRA Nuclear Products Group
Meriden, Connecticut, USA

Abstract

Neutron counters have been used by safeguard inspectors worldwide for over 20 years to

accurately perform Non-Destructive Assays of pure samples of plutonium and uranium. These systems rely on the neutron coincidence counting technique to separate time-correlated fission neutrons from random, uncorrelated neutrons to determine the fissile mass. Impure samples such as mixed-oxide (MOX) scrap materials and salts present a unique problem because they cannot be accurately measured using the standard coincidence technique. Instead, they require an enhanced technique, multiplicity counting, to accurately quantity the fissile content. Test data will be presented in this paper to compare multiplicity and coincidence counting, and to identify sample types that can be assayed using these techniques.

I. Introduction

Several different types of neutron counters that use traditional coincidence counting techniques have been authorized for routine inspection use (AFU) by the IAEA and Euratom, to make accurate verification measurements of pure PuO_2 samples. Such AFU systems include the Inventory Sample Coincidence Counter (INVS), High Level Neutron Coincidence Counter (HLNC), and the Active Well Coincidence Counter (AWCC). Impure plutonium samples, or samples with unknown chemical composition require a more sophisticated technique, multiplicity, to accurately measure the plutonium content. This paper will briefly discuss the standard coincidence and multiplicity techniques, and review test data for MOX standards and scrap material.

II. Discussion of the Techniques

Traditional coincidence counting measures two parameters: the totals neutron rate and the reals or time-correlated rate. The reals rate is determined by subtracting the counts in the Reals+Accidentals (R+A) gate from the Accidentals (A) gate (Figure 1). The gate width for the R+A and A gates varies depending on the counter.

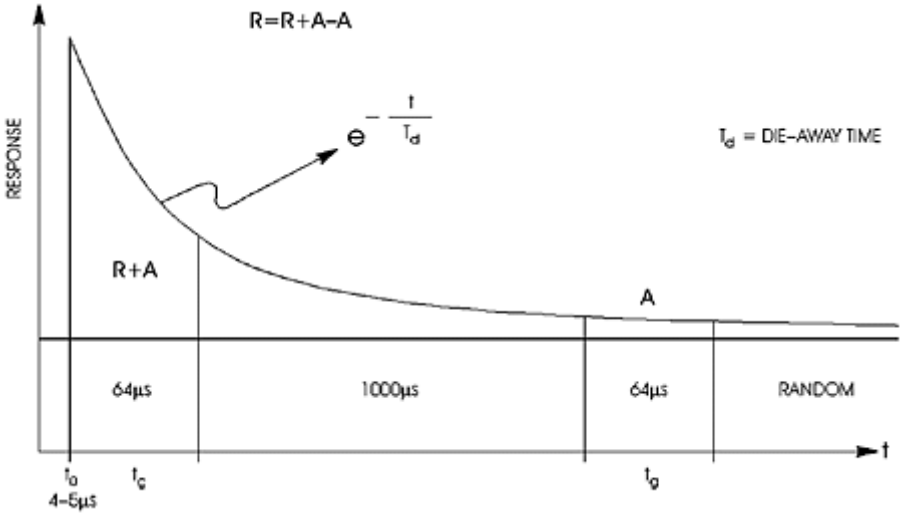

Figure 1.
Simplified diagram showing howthe counter response as a function of time.

With these two measured values, R+A and A, one can determine the plutonium mass and multiplication provided the ratio of random neutrons to coincidence neutrons (e.g. alpha) is known. The equation for alpha is

$$\alpha_0 = \frac{k_1 f_{238} + k_2 f_{239} + k_3 f_{240} + k_4 f_{241} + k_5 f_{242} + k_6 f_{Am241}}{k_7 \left(k_8 f_{238} + f_{240} + k_9 f_{242} \right)}$$

where the isotopic fraction for isotope *i* are represented by f_i, and the values for k_i are known constants for each isotope. For impure plutonium samples it is not valid to assume that the alpha value is known. An enhanced analysis technique, called multiplicity, is required to accurately assay the material.

The multiplicity counting technique has been under investigation at ISPRA and Los Alamos National Laboratory (LANL) for over 10 years to address the problem of NDA for impure samples or samples of unknown chemical composition. The uncertainty in the chemical form of the plutonium leads to an unpredictable level of (?,n) neutron emission. The analysis then requires solution for three unknown values: fissile mass, neutron multiplication, and alpha. Multiplicity counting provides a third measured value allowing a unique solution for the three unknowns. Impure plutonium samples which cannot be analyzed reliably with standard neutron coincidence counting can thus be analyzed with this technique.

Canberra's 2150 multiplicity electronics module is an extension of the IAEA AFU JSR-12 Neutron Coincidence Counter Electronics (NCCE). Where the JSR-12 simply sums the R+A and the A gates, the 2150 multiplicity module records the number of times in 256 scalers that a given number of neutrons is recorded. The R+A and A multiplicity distributions for a MOX standard are listed in Table 1.

Multiplicity	Reals+ Accidentals Counts	Accidentals Counts	Multiplicity	Reals+ Accidentals Counts	Accidentals Counts
0	9605	11373	20	1186436	1039227
1	83045	98659	21	716308	619723
2	375384	434229	22	416231	356067
3	1151735	1312457	23	237073	199777
4	2694830	3010470	24	130075	108311
5	5160160	5662859	25	69755	57013
6	8405588	9069565	26	36816	29950
7	11960226	12676951	27	18642	15155
8	15166519	15827083	28	9647	7555
9	17432091	17882545	29	4728	3710
10	18348968	18537609	30	2154	1637
11	17895079	17798142	31	1014	723
12	16281194	15961957	32	436	344
13	13920981	13436624	33	222	166
14	11235911	10689922	34	108	73
15	8610825	8084184	35	46	41
16	6293619	5824891	36	14	10
17	4405350	4013039	37	9	12
18	2956488	2660166	38	4	6
19	1906225	1691402	39	0	0

Table 1.
Multiplicity Distribution for 1000-sec Measurement of MOX Standard

The multiplicity distribution is used to determine the singles (S), doubles (D) and triples (T) rates as defined by the equations below.

$$S = R_1 / T_M \cdot \left(\frac{m \cdot F}{F_s} \right) = m \cdot F \cdot M \cdot \varepsilon \cdot \bar{\nu}_{s1} \cdot [1 + \alpha]$$

$$D = \frac{R_2}{T_M} \cdot \left(\frac{m \cdot F}{F_s} \right) \cdot f_d = \frac{m \cdot F \cdot M^2 \cdot \varepsilon^2 \cdot f_d}{2} \left\{ \overline{\nu_{s2}} + \frac{M-1}{\overline{\nu_{i1}} - 1} \cdot \overline{\nu_{s1}} \cdot \bar{\nu}_{i2} \cdot (1 + \alpha) \right\}$$

$$T = \frac{m \cdot F \cdot M^3 \cdot \varepsilon^3 \cdot f_t}{3*2} \cdot \left\{ \overline{\nu_{s(3)}} + \left(\frac{M-1}{\overline{\nu_{i1}} - 1} \right) \cdot \left[3 \cdot \overline{\nu_{s2}} \cdot \overline{\nu_{i2}} + \overline{\nu_{s1}} \cdot \overline{\nu_{i3}} \cdot (1 + \alpha) \right] + 3 \left(\frac{M-1}{\overline{\nu_{i1}} - 1} \right)^2 \overline{\nu_{s1}} \cdot (1 + \alpha) \cdot \overline{\nu_{i2}}^2 \right\}$$

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where m is the ^{240}Pu-effective mass, M the multiplication, ε the efficiency, and f_d and f_t the doubles and triples gate fractions. These equations are explained in detail elsewhere.

III. Test Results

The Plutonium Scrap Multiplicity Counter (PSMC), shown in Figure 2, is a commercialized version of a multiplicity counter originally developed by Los Alamos National Laboratory, Group NIS-5, to assay MOX scrap. This PSMC was used to measure well characterized MOX standards of known chemical composition and plutonium isotopics to compare the known alpha technique (traditional coincidence counting) and the multiplicity technique.

Figure 2.
Plutonium Scrap Multiplicity Counter

The PSMC design is optimized for multiplicity counting. The important features include:

- High efficiency - As shown in the equation above, the triples rate is proportional to the efficiency cubed. The PSMC has 80 ^{3}He proportional detectors to obtain an efficiency of ~55%.
- Uniform Response- The PSMC has four rings of ^{3}He proportional detectors arranged to flatten the energy response, and graphite end plugs to flatten the axial response by reflecting neutrons from the dead region of the end plugs back into the ^{3}He detection region.

- Small deadtime - The counter has 19 amplifiers to reduce the deadtime.

Performance characteristics of the PSMC are published in detail elsewhere.

The MULTI code[1] uses both the known alpha technique and the multiplicity technique to analyze the same data set. Results for MOX standards are listed in Tables 2 and 3. Each standard was measured for 10 cycles, 100-sec each for a total of 1000 seconds. The final column in each table is the difference between the declared mass and the measured mass averaged for four 1000-sec assays. This difference gives the accuracy of each verification measurement.

Standard	Totals Rate (error)	Reals Rate (error)	Multiplication	Declared-Measured Mass (%)
1	259080.90 (± 16.3)	56255.19 (± 97.08)	1.07	-1.74
2	171745.28 (± 13.2)	36027.25 (± 63.49)	1.06	-0.37
3	80217.66 (± 8.99)	15441.09 (± 29.35)	1.04	0.78

Table 2.
Known Alpha Analysis of 1000-sec Measurements of MOX Standards

Standard	Singles Rate (Error)	Doubles Rate (Error)	Triples Rate (Error)	Alpha	Multiplication	Declare - Measured Mass (%)
1	255261.18 (± 25.01)	57178.12 (± 0.92)	35137.13 (± 10.28)	0.861	1.080	-0.12
2	170026.8 (± 14.89)	36484.95 (± 0.55)	19154.60 (± 10.98)	0.749	1.057	-1.47
3	79867.96 (± 11.20)	15550.12 (± 0.30)	7003.51 (± 4.38)	0.775	1.037	-1.57

Table 3.
Multiplicity Analysis of 1000-sec Measurements of MOX Standards

The alpha values for MOX standards listed in Tables 2 and 3 are similar to those for pure PuO_2. For both the known alpha and the multiplicity analyses, multiplication ranges from 4-8%. Both techniques give comparable accuracies, demonstrating that either technique can be successfully used for PuO_2 or MOX if the chemical composition and isotopics are well known, and the precision for the triples rate is good.

To test the effect of poor counting statistics, we also made measurements for 10-sec and 100-sec. For standard 1, the declared-measured values ranged from -25% to +38% for 10-sec measurements and 14% for the single 100-sec measurement using the multiplicity technique - primarily due to the inaccuracy in determining the triples rate. For the known alpha technique, the measured values were consistently within 5% of the declared mass for both 10-sec and 100-sec measurements, demonstrating that the known alpha technique is preferred for samples of known chemical composition if the counting statistics are poor.

We also measured nine MOX scrap samples. The scrap results are summarized in Tables 4 and 5 for the known alpha and multiplicity technique, respectively.

MOX Scrap	Assay time (# of	Totals Rate	Reals Rate	Multiplication	Calculated Alpha	Declared- Measured Mass (%)

Sample	cycles * sec/cycle)					
1	10*60	732337.01 (± 53.6)	42895.47 (± 273.1)	1.000	1.125	-177.12
2	10*60	41929.50 (± 12.9)	6337.26 (± 23.7)	1.017	1.115	-8.39
3	10*60	24625.60 (± 8.8)	5300.21 (± 18.6)	1.082	0.953	52.80
4	10*60	6681.34 (± 3.3)	1007.34 (± 3.6)	1.002	0.953	15.02
5	10*60	154906.80 (± 15.2)	12110.57 (± 92.8)	1.000	0.994	-11.21
6	10*60	166580.73 (± 19.2)	22737.46 (± 102.2)	1.000	1.027	-27.01
7	10*60	745320.28 (± 31.3)	203266.67 (± 596.5)	1.153	1.019	0.01
8	10*60	1129615.08 (± 49.8)	147793.92 (± 551.9)	1.000	1.066	-174.85
9	10*60	74901.54 (± 14.6)	19229.96 (± 71.5)	1.097	0.661	13.84

Table 4.
Results of known alpha analysis for MOX scrap material.

MOX Scrap Sample	Singles Rate	Doubles Rate	Triples Rate	Multiplication	Calculated Alpha	Declared-Measured Mass (%)
1	737181.73 (± 53.7)	44110.50 (± 274.7)	30488.8 (± 3236.8)	1.07	27.428	54.5
2	41975.03 (± 12.9)	6364.05 (± 24.0)	2641.48 (± 62.6)	1.026	1.229	-1.72
3	24641.73 (± 8.8)	5316.15 (± 18.7)	2799.73 (± 39.0)	1.057	0.745	46.08
4	6682.60 (± 3.3)	1008.63 (± 3.6)	413.09 (± 5.5)	1.024	1.222	27.20
5	155452.79 (± 15.3)	12263.65 (± 94.1)	5229.93 (± 288.7)	1.024	3.762	14.83
6	167203.64 (± 19.2)	23041.39 (± 105.6)	11749.66 (± 558.5)	1.049	1.916	11.09

7	750184.31 (± 31.3)	208886.08 (± 612.7)	171860.11 (± 2786.7	1.133	0.814	-13.64
8	1131135.73 (± 48.8)	150568.91 (± 552.5)	126914.87 (± 11692.4)	1.122	6.138	23.55
9	75041.30 (± 14.7)	19367.05 (± 72.5)	11142.81 (± 197.0)	1.071	0.496	2.23

Table 5.
Results for multiplicity analysis for MOX scrap material.

For the known alpha technique (Table 4), the differences between the declared and measured masses are significantly worse than those for the MOX standards (Table 5). This is as expected because the chemical composition and the declared masses of the scrap are not as well known as those for the standards. For most of the verification measurements for MOX scrap material listed in Tables 4, the calculated alpha values were larger than expected for pure MOX, indicating a third unknown. Therefore, the known alpha technique is not valid for these samples. For samples 1, 5, 6, and 8 of the known alpha analyses, the multiplication values are 1.000. The MULTI software sets multiplication to 1.000 if the calculated value is <1, indicating that alpha is larger than expected - further proof that the known alpha technique is not valid.

For the multiplicity technique (Table 5), the differences are smaller than the known alpha technique. In particular, samples 1 and 8 which had the largest difference using the known alpha technique are significantly improved with the multiplicity technique. MOX scrap sample 1 gave an alpha value of 27.43 but the error in the reported mass was very large, indicating a questionable verification result. We measured the isotopics of the sample using Multi-Group Analysis Code (MGA) to confirm the declared values to minimize the error from the isotopics. The measured results showed good agreement with the declared values. The high singles rate compared to the doubles rate for sample 1 in Table 5 indicates that the sample could contain fluoride. Other possible explanations include the presence of unknown alpha emitters or an unexpected chemical form for the plutonium. MOX scrap sample 1 was re-assayed for 12 runs of 600 sec each to improve the precision of the triples rate. The new assay gave an alpha of 15.923. The difference between the declared and measured mass was 22.83%. This test suggests that samples with large alpha values that give poor results in the PSMC in 600 sec count times can be re-assayed for longer count times to improve the accuracy of the multiplicity analysis. The count time per run should also be increased to improve the precision of the triples rate. This measurement also demonstrated that the multiplicity electronics and analysis can provide useful results for alpha values in excess of 10. There was not sufficient time to repeat all of the other scrap samples at longer count times.

IV. Conclusion

The test results lead to the following conclusions.

- If the value of alpha is known (e.g. the isotopics and chemistry are well known) then it is possible to perform a correction for the multiplication effects with only two measured values.
- If there are three unknowns, fissile mass, multiplication, and alpha, multiplicity analysis is required. When the (?,n) emission rate is uncertain due to poorly known isotopics or uncertain chemistry the standard known alpha technique fails. By adding the third measured value it is possible to determine the (?,n) emission rate. With the measured value for (?,n) neutron emission, the plutonium mass can be measured without complete knowledge of the chemical form, but there are restrictions on the count rates. Assay of MOX scrap with alpha values as large as 15 were demonstrated using the PSMC. For higher ratios the (?,n) counting statistics become worse and the accuracy of the measurement suffer.