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Author(s):

M. S. Krick
D. G. Langner
D. W. Miller
J. R. Wachter
S. S. Hildner

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THERMAL-NEUTRON MULTIPLICITY COUNTER MEASUREMENTS*

M. S. Krick, D. G. Langner, D. W. Miller,
J. R. Wachter, and S. S. Hildner
Los Alamos National Laboratory
Los Alamos, NM 87545 USA

ABSTRACT

An in-plant thermal-neutron multiplicity counter was designed and built to assay plutonium samples for which the relative (α, n) neutron yield and the neutron multiplication are not known. Such samples are difficult to assay with conventional neutron coincidence counting techniques. We have characterized this counter with various ^{252}Cf sources and plutonium samples to study the neutron detection efficiency, electronic dead-time, and neutron die-away properties of the detector and associated electronics. We used a new shift-register electronics module that features 256 channels for multiplicity counting. Preliminary reference parameters have been obtained for performing assays with the multiplicity counting system. We measured a variety of plutonium materials with the in-plant counter in the Los Alamos Plutonium Facility. The materials included samples with high neutron multiplication and samples with high (α, n) neutron yields. Preliminary assay results are presented.

INTRODUCTION

The in-plant thermal-neutron multiplicity counter has been used for a measurement exercise in the Los Alamos Plutonium Facility to assay plutonium in the form of oxide, metal, and process material that has a high neutron yield from (α, n) reactions. The multiplicity counter provides three measured quantities for the determination^{1,2} of the three principal unknowns: the effective ^{240}Pu mass ($^{240}\text{Pu-eff.}$), the neutron multiplication (M), and α , the ratio of (α, n) neutrons to spontaneous-fission neutrons.

The solution of the multiplicity equations for the three unknowns will be referred to as three-parameter analysis. The data were also analyzed with a two-parameter analysis, which uses two of the three multiplicity

equations and is the same as the Ensslin multiplication-correction procedure³ with Bochnel's equations⁴ for the multiplied source moments. One-parameter analysis simply uses a calibration curve with the usual coincidence rate plotted vs $^{240}\text{Pu-eff.}$

MEASUREMENT SYSTEM

Detector

The in-plant thermal-neutron multiplicity counter has been described in previous publications.⁵⁻⁷ Cross-sectional drawings of the detector are also shown elsewhere in these proceedings.⁸

Electronics

The prototype multiplicity shift-register circuit (the MSR4)⁹ operates with a 4-MHz clock and is packaged in a double-wide nuclear instrumentation module. The MSR4 is a conventional shift-register coincidence circuit and in addition is a multiplicity-sorting circuit. Two multiplicity distributions are measured: one for the real-plus-accidental ($\langle R + A \rangle$) coincidence gate and one for the accidental ($\langle A \rangle$) coincidence gate. An example is shown in Table I, where the $\langle R + A \rangle$ and $\langle A \rangle$ distributions are shown for a plutonium oxide sample. The example shows the number of times that each gate contained a specified number of neutron pulses (the gate multiplicity). In general, multiplicity analysis utilizes the moments or the multiplets of the $\langle R + A \rangle$ and/or the $\langle A \rangle$ distributions.

Software

No analysis is done in the multiplicity module; the raw data are transferred across a serial interface to an IBM-compatible personal computer. The data for this exercise were collected with a special code written for research purposes, but a code for routine use is now available. The new code is named MULTI and is one in a series of neutron coincidence counting codes developed by the Safeguards Assay Group at Los Alamos. The MULTI code collects, stores, and retrieves raw data; does quality control tests; and calculates results and errors.

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TABLE I. Sample Multiplicity Distribution

Multiplicity	Counts	
	R + A	A
0	4 863 880	5 708 122
1	16 014 347	17 977 225
2	27 615 893	29 696 698
3	33 191 194	34 213 111
4	31 244 355	30 927 348
5	24 535 179	23 349 181
6	16 706 604	15 317 903
7	10 161 183	8 979 892
8	5 604 155	4 785 248
9	2 853 120	2 359 023
10	1 356 495	1 082 439
11	606 426	469 055
12	257 046	192 488
13	103 926	75 358
14	40 246	28 536
15	15 106	10 416
16	5 522	3 736
17	1 872	1 193
18	702	440
19	216	136
20	78	52
21	21	9
22	10	5
23	6	1
24	2	0
25	0	0

Technique

The $\langle R + A \rangle$ and $\langle A \rangle$ multiplicity distributions are converted to deadtime-corrected moments using the procedure developed by Dylewski.¹⁰ The moments of the reals ($\langle R \rangle$) distribution are calculated from the $\langle R + A \rangle$ and $\langle A \rangle$ moments and the background is sub-

tracted. The first three moments of the reals distribution are referred to conveniently as the singles (S), doubles (D), and triples (T). S and D are the same as totals and reals in conventional coincidence counting. The moments equations of Hage and Cifarelli¹¹ and Boehnel⁴ are solved to obtain M , α , and ^{240}Pu -eff. from S , D , and T . The solution of the moments equations does not involve sample-specific information, that is, there is no calibration in the manner of conventional neutron coincidence counting. The solution does require four detector parameters and several nuclear data constants.

The four detector parameters are the neutron detection efficiency, the electronic deadtime, the doubles gate fraction, and the triples gate fraction. The gate fractions take into account the fraction of the neutron die-away time that the coincidence gates are open.

There are several assumptions implicit in the moments equations. The two most important assumptions are that the neutron detection efficiency is independent of neutron energy and that the probability of a neutron inducing a fission is the same for all neutrons, that is, the neutron multiplication is a constant. These conditions are never quite satisfied and thus contribute to assay errors.

MEASUREMENTS

Californium-252

The four detector parameters were determined using ^{252}Cf sources, one of which has an absolute yield determined by the National Institute of Standards and Technology. The neutron detection efficiency is 58.3%.

The measured triples/doubles ratio (T/D) for ^{252}Cf should be independent of the neutron yield, but the observed ratio decreases with increasing count rate as a result of electronic deadtime. The multiplicity distributions were corrected for deadtime by using Dylewski's procedure. The deadtime was selected so that after deadtime correction, T/D was constant for all ^{252}Cf sources up to a singles count rate of 250 000 counts/s. The deadtime is 90 ns.

The doubles and triples gate fractions were obtained from the reference ^{252}Cf source using the three moments equations, the known neutron yield, and the known ^{252}Cf source multiplicity distribution. The doubles gate fraction is 0.4377 and the triples gate fraction is 0.1945.

Plutonium

Twenty plutonium samples were measured with the multiplicity counter at the Los Alamos Plutonium Facility. The sample IDs and material types are listed in Table II. Five were oxides, five were metals, and ten were high- α materials. Two of the oxides (CALEX1 and STD40) were well-categorized standards. For all the other samples, the reference ^{240}Pu -eff. values were obtained from calorimetry combined with gamma-ray isotopic measurements. The gamma-ray isotopic measurements produced a specific power to a typical accuracy of 0.5% to 2% and a ^{240}Pu -eff. mass to a typical accuracy of 3% to 7%. Most of the samples had about 6% ^{240}Pu .

The high- α XBLs and XBLC materials are produced in an electrorefining process. They both contain KCl and NaCl salts. The XBLs materials have a high ^{241}Am content, whereas the XBLC materials have a low ^{241}Am content. The XBLP materials are produced in a molten-salt extraction process; they contain salts and have a high ^{241}Am content.

The measured samples were radially centered in the sample cavity of the detector and about 10 cm above the bottom of the cavity. The samples were usually measured for 1500 s (15 runs of 100 s each) twice. One of the oxide standards (STD40) was measured frequently for quality control. We also made frequent background measurements.

RESULTS

General

The background count rates were very small compared to the sample count rates. No inconsistencies were found in repeat measurements of 15 x 100 s each. The measurements of STD40 showed that the detector was stable throughout the exercise. The deadtime- and background-corrected singles, doubles, and triples rates for the samples are given in Table II.

Multiplication and α

Multiplication and α values are produced by the three-parameter analysis. Multiplication and α can also be obtained from a two-parameter analysis if the ^{240}Pu -eff. mass is known. Reference values for the multiplication and α were obtained using the ^{240}Pu -eff. mass from the calorimeter and gamma-ray isotopics measurements. These two sets of multiplication and α values are listed for comparison in Table III. Note the variations of these quantities with sample type.

TABLE II. Count Rates for the Assay Samples

Sample ID	Type	Count Rate (counts/s)		
		Singles	Doubles	Triples
CALEX1	Oxide	29 115	5 295	2 805
LZB205-RM600	Oxide	103 920	19 607	12 597
LZB205-RM595	Oxide	102 557	21 186	14 797
LZB205-RM585	Oxide	89 859	22 534	17 526
STD40	Oxide	117 979	21 547	10 308
PMA2380	Metal	16 521	5 349	3 796
PMA1439	Metal	29 624	10 651	9 387
PMA2379	Metal	30 950	11 500	10 208
METAL1	Metal	178 340	183 159	615 717
PMA1440	Metal	223 920	223 949	704 811
XBLS97	High- α	20 670	366	96
XBLS96	High- α	33 590	743	220
XBLP260	High- α	112 264	1 050	265
XBLP272	High- α	83 756	1 013	271
XBLP268	High- α	82 752	1 070	323
XBLS91	High- α	37 017	134	47
XBLS94	High- α	64 680	1 397	418
XBLP265	High- α	136 792	1 852	544
XBLP280	High- α	86 234	1 660	492
XBLC2163A	High- α	163 657	3 647	1 214

Plutonium-240-eff. Assay Results

The reference ^{240}Pu -eff. masses and the three-parameter ^{240}Pu -eff. mass assay results are shown in the last three columns of Table III. The error of the mass difference is primarily a result of neutron counting statistics for the high- α samples and the oxide standards; for the LZB oxides and the metal samples, the error is primarily a result of the gamma-isotopic ^{240}Pu -eff. mass error.

One of the high- α samples (XBLS91) had grossly inconsistent assay masses as determined by the calorimeter and neutron measurements. Because the neutron data are internally consistent, the calorimeter value is in doubt; although this sample is included in the data tables, it was excluded from the calculations of average results.

TABLE III. Comparison of Reference and Assay Values								
Sample ID	Type	Multiplication		α		^{240}Pu -eff. Mass (g)		(Assay-Reference)/Reference (%)
		Reference	Assay	Reference	Assay	Reference	Assay	
CALEX1	Oxide	1.096	1.103	0.881	0.964	23.76	22.63	-4.8 $\pm$ 0.4
LZB205-RM600	Oxide	1.141	1.140	1.311	1.308	66.29	66.42	0.2 $\pm$ 3.4
LZB205-RM595	Oxide	1.154	1.158	1.149	1.185	69.56	67.84	-2.5 $\pm$ 3.5
LZB205-RM585	Oxide	1.181	1.186	0.804	0.850	70.93	68.97	-2.8 $\pm$ 3.8
STD40	Oxide	1.079	1.085	0.718	0.772	107.00	103.20	-3.6 $\pm$ 0.2
PMA2380	Metal	1.177	1.170	0.188	0.152	19.86	20.62	3.8 $\pm$ 7.4
PMA1439	Metal	1.239	1.223	0.289	0.181	31.19	33.99	9.0 $\pm$ 5.2
PMA2379	Metal	1.212	1.224	0.105	0.154	38.86	36.72	-5.5 $\pm$ 5.4
METAL1	Metal	1.741	1.790	(-0.023) ^a	0.180	176.43	142.07	-19.5 $\pm$ 3.5
PMA1440	Metal	1.724	1.751	(-0.018) ^a	0.089	222.50	197.52	-11.2 $\pm$ 2.9
XBLS97	High- α	1.006	1.005	14.43	13.50	2.24	2.39	6.7 $\pm$ 6.2
XBLS96	High- α	1.007	1.012	11.22	12.80	4.59	4.05	-11.8 $\pm$ 9.5
XBLP260	High- α	1.006	1.002	32.23	29.21	5.65	7.15	26.6 $\pm$ 25.3
XBLP272	High- α	1.005	1.004	21.19	21.25	5.84	6.31	8.1 $\pm$ 15.8
XBLP268	High- α	1.005	1.009	22.87	22.23	6.33	5.14	-18.8 $\pm$ 20.4
XBLS91	High- α	(0.960) ^a	1.006	8.62	315.00	6.74	0.20	-97.0 $\pm$ 5.2 ^b
XBLS94	High- α	1.015	1.012	14.83	13.80	6.77	7.42	9.6 $\pm$ 8.8
XBLP265	High- α	1.010	1.009	24.01	25.60	9.11	9.61	5.5 $\pm$ 26.4
XBLP280	High- α	1.009	1.011	14.74	16.25	9.13	8.85	-3.1 $\pm$ 11.1
XBLC2163A	High- α	1.014	1.019	13.84	18.97	18.29	15.52	-15.1 $\pm$ 12.2

^aNonphysical values are in parentheses.
^bNot included in the average results in Table IV.

TABLE IV. Average Assay Biases for Three-Parameter Analysis	
Sample Type	Bias (%)
Oxide	-2.7 $\pm$ 0.8
Metal	-4.7 $\pm$ 5.1
High- α	0.9 $\pm$ 4.8
All samples	-1.5 $\pm$ 2.6

The average results for the three-parameter analyses are shown in Table IV, where the unweighted average biases and sample standard deviations are given for oxides, metals, high- α samples, and all samples as one group.

DISCUSSION

The three-parameter analysis is based on detector parameters measured with ^{252}Cf sources and contains no sample-dependent calibration parameters and no correction factors. The analysis procedure is identical for oxides, metals, and high- α samples. The ^{240}Pu -eff. assay masses tend to be biased negative and the reason for this is under investigation. The absolute neutron yield of the reference ^{252}Cf source is known to 1.4% (1 standard deviation). Also, a 1% error in the doubles gate fraction corresponds to an assay mass error of 2% to 5%, depending on the sample. Thus, detector parameters could account for part of the bias. In a practical assay situation, such a bias can simply be normalized out for a particular material type by referencing to a standard plutonium sample, but this introduces an undesirable, sample-dependent, calibration parameter.

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