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AUTHOR(S) D. G. Langner, N. Dytlewski, and M. S. Krick

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Los Alamos, New Mexico 87545

PYROCHEMICAL MULTIPLICITY COUNTER DEVELOPMENT*

D. G. Langner, N. Dytlewski,** and M. S. Krick
Los Alamos National Laboratory
Safeguards Assay Group, N-1
Los Alamos, NM 87545 USA

ABSTRACT

Impure plutonium-bearing materials from pyrochemical processes often display both significant self-multiplication and variable (α,n) reaction rates. Standard neutron coincidence counting techniques usually fail to accurately measure these materials. Neutron multiplicity counters measure the third moment of the neutron multiplicity distribution and thus make it possible to deduce the fertile plutonium mass of a sample even when both the self-multiplication and the (α,n) reaction rate are unknown. A multiplicity counter suitable for measuring pyrochemical materials has been designed and built. This paper describes the results of characterization studies for the new counter. The counter consists of 126 helium-3 tubes arranged in 4 concentric rings in a polyethylene moderator; the average spacing between the tubes is 1.59 cm. The end plugs for the counter are made of graphite, and the 24.1- by 37.5-cm sample cavity is cadmium lined. The counter consists of two distinct halves from which the neutron counts are summed. The counter is capable of operation in either a freestanding mode with the two halves coupled together by an external cabinet or in a glove-box mode with the two halves placed around a glove-box well and then mated. For a ^{252}Cf source centered in the sample cavity, the measured efficiency of the new multiplicity counter is 57.7% and its die-away time is 47.2 μs.

INTRODUCTION

Conventional neutron coincidence counting techniques attempt to deduce the fertile mass of a plutonium sample from the measured first and second moments of the sample's neutron multiplicity distribution. In addition to the sample's fertile content, however, this distribution depends on the self-multiplication of neutrons in the sample and on the (α,n) neutron rate. Thus, conventional, two-parameter coincidence counting techniques can only be successful when measuring pure materials or materials whose (α,n) neutron rate or self-multiplication are known.[1]

Neutron multiplicity counters also measure the third moment of the neutron multiplicity distribution and can assay impure plutonium-bearing materials that are not so well characterized.[2,3] To successfully measure the third moment and obtain accurate assays using a multiplicity counting technique, a thermal neutron detector must be carefully designed to have a high detection efficiency, low die-away time, and a detection efficiency that is as invariant as possible when there are variations in sample geometry and in the neutron energy spectrum.[4]

At Los Alamos, we have designed and built a counter to meet the above criteria and to be suitable for in-plant measurement of a wide range of impure materials such as those encountered in pyrochemical processes. This new counter, dubbed the pyrochemical multiplicity counter, was also designed to be capable of both at-line and in-line measurement and to accommodate a wide variety of sample containers. In the first phases of the design and conception of this counter, computer simulations were performed to optimize the placement of its ^3He tubes and the design of its end plugs relative to the above-mentioned criteria. The results of these simulations are described in Ref. 5. This paper reports on the construction of this new multiplicity counter and characterization studies performed on it.

COUNTER CONFIGURATION

Figure 1 is a conceptual diagram of the pyrochemical multiplicity counter. It consists of 126 helium-3 tubes with 71-cm active-lengths arranged in 4 concentric rings and embedded in a polyethylene body. The tube spacing is approximately 1.59 cm to maximize efficiency but minimize die-away time. The sample cavity is a large 24.1-cm wide by 37.5-cm high and is optimized for cans up to 20-cm wide and 36-cm high. The sample cavity is lined with 0.081 cm of cadmium and the end caps are made of graphite. The outer rings of tubes are protected from room background by a 10.2-cm polyethylene shield, and the design allows for the addition of another 10.2-cm polyethylene shield above the junction box for in-line applications.

To allow the counter to be used in a free-standing, at-line mode or wrapped around a glove-box well for in-line measurements, we built it in two halves as shown in Fig. 2. There are two high-voltage junction boxes, one for each half of the counter as shown in Fig. 3. In each junction box there are 18 AMPTEK preamplifiers to handle the load of the 63 tubes. These preamplifiers are distributed nonuniformly from ring to ring to accommodate the changes in detection efficiency. The innermost ring, which consists of 11 tubes per half, is serviced by 6 AMPTEKs. The next ring out, which has 14 tubes per half, is serviced by 5 AMPTEKs.

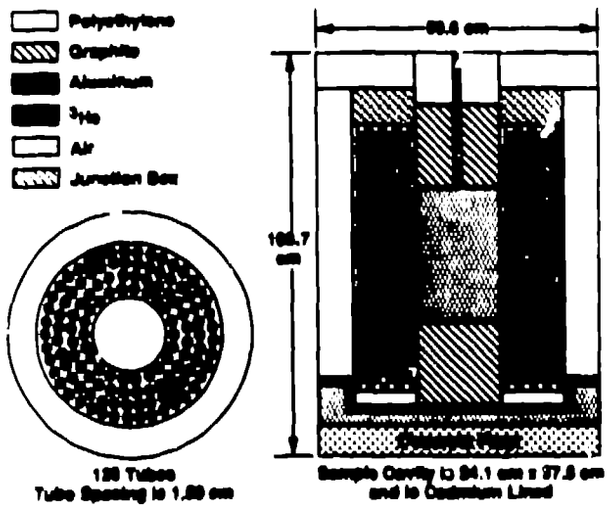

Fig. 1. Schematic of the pyrochemical multiplicity counter.

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**Visiting Scientist from the Australian Nuclear Science and Technology Organisation, Private Mail Bag, Menai NSW 2234, Australia

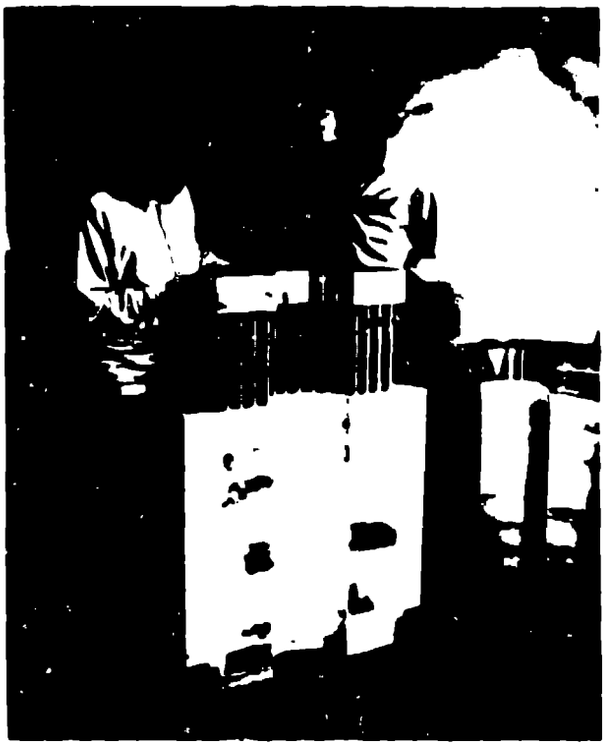

Fig. 2. The two halves of the pyrochemical multiplicity counter.

Fig. 3. The pyrochemical multiplicity counter's junction box showing the layout of the AMPTEK preamplifiers.

The last 2 outer rings, which have 17 and 21 tubes per half, are serviced by 4 and 3 AMPTEKs, respectively. The high voltage of 1680 V and the 5-V input to the AMPTEKs is supplied separately to each half of the junction box. The output signals from each half-ring are then brought out separately and summed so that the relative responses of the rings to changes in sample geometry and make-up can be examined.

DETECTION EFFICIENCY AND SPATIAL CHARACTERISTICS

Simulations of the pyrochemical counter's response to a ^{252}Cf source placed in the center of the sample chamber predicted that the counter would have a detection efficiency of 57.0%, an exponential die-away behavior, and a die-away time of 49.4 μs. The measured efficiency of the counter is 57.7%, and its measured die-away time is 47.2 μs. The measured die-away response of the counter is shown in Fig. 4 together with an exponential fit. The differences between the measurements and the calculations are consistent with slight design changes that were made to accommodate the split-counter geometry. These changes included moving one tube from the third ring to the fourth ring and moving the tubes slightly closer together than the specified 1.59 cm to allow for more space between the outermost tubes and the junction box wall to eliminate the possibility of high-voltage breakdown. The calculations were not repeated after these slight changes because we did not expect them to significantly affect the results.

The spatial response of the counter to a ^{252}Cf source centered in the sample chamber and moved axially and radially is shown in Figs. 5 and 6. Several calculated values are overlaid for comparison. In the center of the sample chamber, the pyrochemical counter's response is very uniform. The agreement between the calculations and the measured values is within the precision of the calculations. The greater decrease in the axial response of the counter near the top of the sample chamber compared to its behavior at the bottom is due to neutron leakage through the junction box.

SAMPLE MEASUREMENTS

A figure-of-merit study performed by Ensslin et al.[4] has shown that the assay precision for multiplicity counters depends on the detector's efficiency and die-away time; on a sample's plutonium content, self-multiplication, and (α,n) neutron rate; and on counting parameters including the coincidence gate width, predelay, and count time. Using Ensslin's method, the assay precision was calculated for oxide samples measured in the pyrochemical counter for

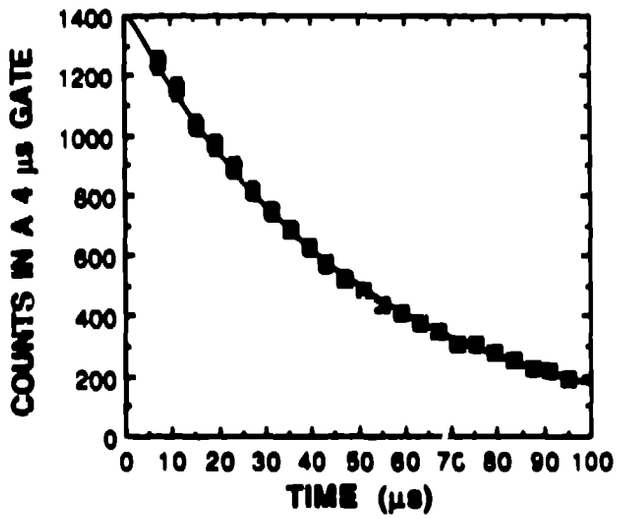

Fig. 4. The measured die-away response of the pyrochemical multiplicity counter to a ^{252}Cf source placed in the center of the sample chamber.

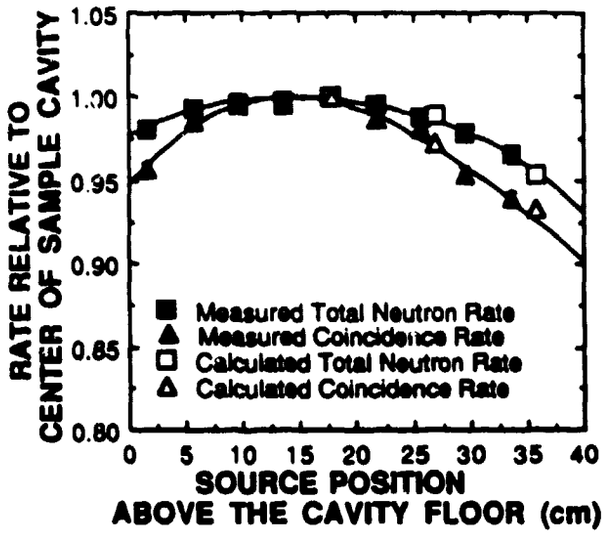

Fig. 5. Axial response of the pyrochemical multiplicity counter to a ^{252}Cf source.

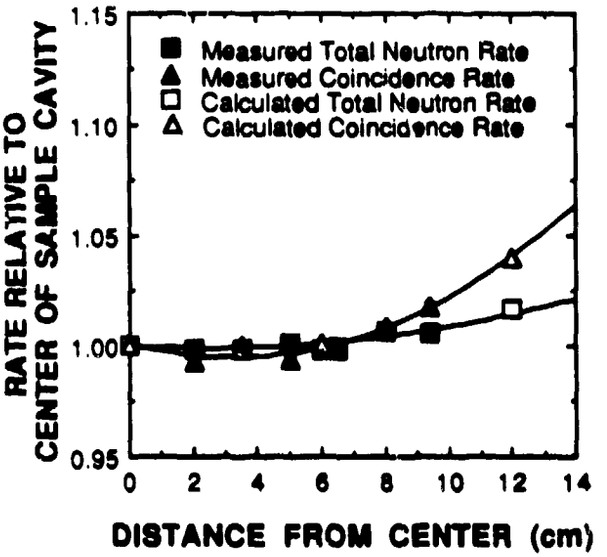

Fig. 6. Radial response of the pyrochemical multiplicity counter to a ^{252}Cf source.

2000 s using a coincidence gate width of 32 μs and a predelay of 3 μs. These calculations, shown graphically in Fig. 7, predict that the pyrochemical counter should measure oxides whose ^{240}Pu-effective mass is less than 300 g to a precision of 1% or less. The ^{240}Pu-effective mass of a sample is the equivalent amount of ^{240}Pu, which would produce the same coincidence rate as the sample. A sample containing other even isotopes of plutonium will have a ^{240}Pu-effective mass greater than its actual ^{240}Pu content.

Similar calculations were performed to predict the assay precision of the pyrochemical counter as a function of (α,n) neutron rate. For this purpose, the ratio α of (α,n) neutrons to spontaneous-fission neutrons in a sample is used. This calculation was done using the same parameters as above except for a small sample containing the equivalent of 0.56 g of ^{240}Pu and having self-multiplication of 1%.

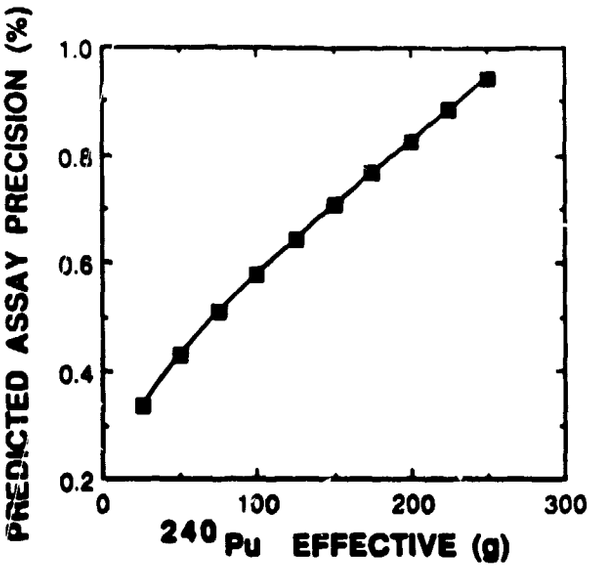

Fig. 7. Predicted assay precision for plutonium oxide samples measured in the pyrochemical multiplicity counter.

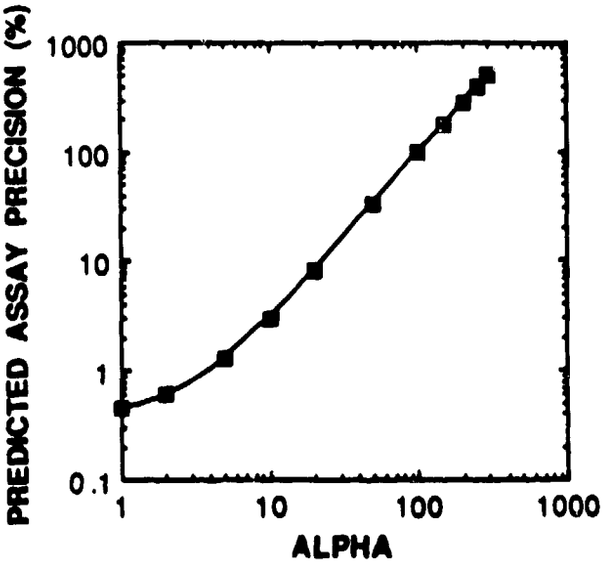

Fig. 8. Predicted assay precision for the pyrochemical multiplicity counter for a small plutonium sample as a function of (α,n) neutron rate.

This size sample was chosen to match a set of small samples that was later assayed in the pyrochemical counter. Figure 8 shows the predicted assay precision. From these calculations, assays with precisions of 5% or less should be possible in 2000 s for samples with α's less than about 15.

Eight small samples each having less than 15-g total plutonium but having a variety of matrices were assayed in the pyrochemical counter using a multiplicity circuit capable of processing multiplicities of up to 32 and counting parameters as used in the calculations above. Pertinent data concerning these samples are given in Table I. The analysis of the measured multiplicity distributions from these samples used the moments method developed by Boehnel[6] and Cifarelli and Hage[7] and included a deadtime correction based on a scheme derived by Dytlewski.[8] The assay calibration

Table I. Multiplicity Assay Results for Small Plutonium Samples Obtained with Pyrochemical Multiplicity Counter

Sample Type	Matrix	Total Plutonium (g)	$^{240}Pu_{Eff}$ (g)	Multiplicity Assay Results α	M	Assay/ Reference	"Known M" Assay/ Reference
Oxide	None	10.0	0.564	1.60	1.008	0.991	1.010
Oxide	Aluminum	10.0	0.564	6.12	1.006	1.002	1.054
Oxide	Magnesium oxide	10.0	0.564	11.7	1.006	1.040	1.172
Oxide	Silicon	10.0	0.564	2.56	1.003	0.994	0.981
Oxide	Boron	10.0	0.565	276.0[a]	1.093	6.227	7.154
Fluoride	Fluorine	10.0	0.563	159.0[a]	1.052	4.758	5.044
Metal	None	10.0	0.529	0	1.010	1.000	1.000
Oxide	None	12.4	0.700	0.161	0.997	1.012	0.958
				Average for α less than 7		0.999	1.001
				Standard Deviation		3.6%	0.80%

[a]The α obtained from the multiplicity assay for these samples is negative and therefore anomalous. The α reported for these samples was obtained from the total neutron rate by assuming that $M = 1$. The self-multiplication results are also suspect.

was based on values for detection efficiency and gate fractions derived from measurements of the sample containing pure plutonium metal. We assumed that α is zero for this sample and its self-multiplication is 1.01. This last value was based on previous measurements of the samples in another multiplicity counter.

The multiplicity assays of these samples in the pyrochemical counter can be considered successful for all but the samples containing boron and fluorine. The assay of the sample containing magnesium oxide was high by 4%, but this is within the predicted precision based on the moderately large α for this sample. For the samples containing boron and fluorine, the assay values for α were negative and therefore anomalous. Also the multiplication results are much too high for samples this small. However, α can be estimated to within a few percent from the measured total neutron rate for these samples given the known ^{240}Pu-effective content, the known detector efficiency, and the assumption that the multiplication is one. For these samples the estimated α is very large and the predicted precision is several hundred percent. Multiplicity analysis, therefore, should not be successful for samples whose α's are so large.

Conventional two-parameter assay would normally not be attempted for these samples because both multiplication and α are varying. However, because the self-multiplication of these samples is near one, a two-parameter assay of these samples was performed for comparison by assuming that the multiplication was one. This assay was accomplished using the measured first and second moments and a calibration based on the small metal sample. The assay results using this "known-M" technique are given in Table I and are plotted with the results obtained using the multiplicity analysis in Fig. 9. The boron sample and the fluorine sample results are not plotted. For the samples whose α's are < 7, the two-parameter assay results are accurate to 3.6% (1σ) whereas the multiplicity assays are accurate to 0.8%. No bias is evident in the assay results for this set of samples.

CONCLUSIONS

The characterization results for the pyrochemical multiplicity counter demonstrate that the design goals identified

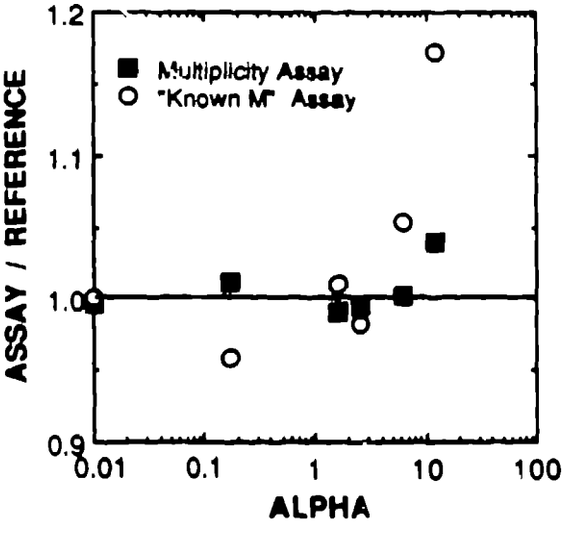

Fig. 9. A comparison of the multiplicity assay results obtained with the pyrochemical multiplicity counter for small, impure plutonium-bearing samples to results obtained using a conventional "known-M" assay technique.

for the counter have been achieved. Assays accurate to 1% are possible for samples whose ratio of (α,n) neutrons to spontaneous-fission neutrons is $\lesssim 7$.

Future work will concentrate on measuring larger samples and samples more representative of pyrochemical process materials. In addition, the response of the various rings to different samples will be studied to ascertain if variations in ring response can be used to detect sample-to-sample changes in the detection efficiency of the counter that are due to such factors as changing sample composition and sample geometry.

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