LA-UR 91-1569

LA-UR--91-1569

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Los Alamos National Laboratory is operated by the University of California for the United States Department of Energy under contract W-7405-ENG-36

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SUBMITTED TO: European Safeguards Research and Development Corporation
(ESARDA)
Avignon, France
May 14-16, 1991
(Full Paper)

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FORM NO 836 R4
ST NO 2629 5/81

NEUTRON MULTIPLICITY COUNTER DEVELOPMENT*

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Abstract

We have designed and built two multiplicity counters to test the usefulness of multiplicity counting for the safeguards assay of plutonium-bearing materials. The first prototype counter has been characterized and a variety of plutonium-bearing materials have been measured with it. Assays accurate to ~0.7% have been obtained for both pure and impure plutonium oxide samples in reasonable measurement times. Assays accurate to ~5% have been obtained for metal samples. A second multiplicity counter has been designed using experience gained from the first as well as Monte Carlo simulations. The second counter was designed to be more suitable for in-plant measurement of pyrochemical process materials. This paper presents the results of characterization studies of the two instruments.

1. Introduction

A conventional passive neutron coincidence assay of plutonium measures the first and second moments of the neutron multiplicity distribution of a sample and deduces from them the sample's fertile mass. The problem with this technique is that there are actually three variables in the measurement problem: the sample mass, the (α,n) neutron rate of the sample, and the multiplication of neutrons by the sample. Thus, conventional coincidence techniques only provide accurate assays for pure materials; for samples whose impurities are known well enough to compute the ratio, α, of their (α,n) neutron rate to their spontaneous-fission rate; or for impure samples whose self-multiplication is known /1/.

The goal of neutron multiplicity counting is to obtain a third quantity from the multiplicity distributions and to deduce all three variables from measured quantities. Our first assays using multiplicity counting and a conventional thermal-neutron detector were obtained from measurements performed in a modified, active-well counter using specialized shift-register electronics designed and built just for the purpose /2/. These assays were accurate to ~2% (1σ) for a variety of pure and impure materials. However, very long measurement times were necessary to obtain this accuracy for large samples with an α greater than about 0.5 because of the large statistical error in the measured triples to doubles ratio.

Assays of this accuracy can be obtained in more reasonable measurement times if the neutron counter is designed to have a very high detection efficiency relative to most thermal-neutron counters used for safeguards measurements and a low die-away time. Short electronic deadtime is also essential if accurate deadtime corrections are to be performed. This paper reports on two thermal-neutron counters that have been designed and built at Los Alamos to achieve the goal of accurate multiplicity assays in reasonable measurement times. The first prototype counter was built to study the effects of counter design on multiplicity assay. It was designed to be used with multiplicity-sorting electronics that could sort multiplicities up to seven and a moment analysis technique that greatly reduces the measurement times needed to obtain good statistical precision for large samples. Using the experience gained from this first counter, we designed and built a second counter to be more applicable to facility measurements.

2. The Dual-Mode Multiplicity Counter

The first multiplicity counter consists of 130, 50.8-cm-active-length ^{3}He tubes. A schematic diagram of this counter is shown in Fig. 1. In the counter's initial configuration, each of the ^{3}He tubes was wrapped in a 0.7-cm-thick sleeve of high-density polyethylene followed by an outer, 0.081-cm-thick sleeve of cadmium. Each wrapped tube was then placed in the aluminum core as shown in the diagram. From the Monte Carlo design calculations we expected this configuration of the counter to have a low detection efficiency but a very low die-away time. The short die-away time was essential so that short coincidence-gate widths could be used without loss of counting precision. Short gate widths were necessary so that the multiplicity circuit would not overflow for large samples. The counter was called the dual-mode counter because it was also designed to operate in a high-efficiency mode with the cadmium sleeves removed.

Early characterization studies of the counter in its low-efficiency mode (cadmium sleeves in place) revealed a detection efficiency of 17% for a ^{252}Cf source placed in the center of its 16.5-cm-wide by 25.4-cm-high sample chamber and a

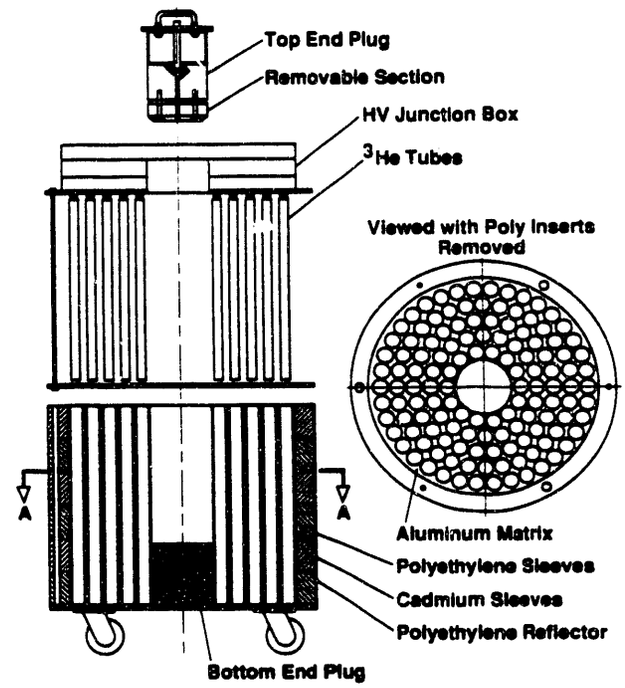

Fig. 1. Schematic of the dual-mode multiplicity counter.

*This work is supported by the US. Department of Energy, Office of Safeguards and Security.
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die-away time of 11.8 μs. In the high-efficiency mode, the measured detection efficiency was 53% for ^{252}Cf and its die-away time was 57 μs.

Using assay variance as a figure-of-merit for multiplicity counters, Ensslin et al. /3/ have written a program that predicts the statistical accuracy of a multiplicity assay as a function of a detector's efficiency and its die-away time; a sample's effective ^{240}Pu content, self-multiplication, and (α,n) neutron rate; and the measurement count time. They have calculated the precision of assays performed by this prototype counter for samples whose α is less than 5 and whose ^{240}Pu-effective content is between 7.6 and 143 g. These calculations predicted that the dual-mode counter will perform more accurate assays in the high-efficiency mode than in the low-efficiency mode for samples of this type. In the high-efficiency mode, however, the shift-register multiplicity circuit had to be upgraded to handle higher multiplicities. In Ref. 3, Ensslin et al. report on measurements made with this counter in the low-efficiency mode using the original multiplicity circuit that could sort multiplicities up to seven. Measurements made in the high-efficiency mode with a new multiplicity circuit capable of sorting multiplicities up to 32 are reported here.

Figures 2 and 3 show the spatial response of the dual-mode counter in the high-efficiency mode measured with a ^{252}Cf source centered in the sample chamber and moved axially and radially. This detector's response is uniform in the central region of the cavity, decreases near the end plugs, and increases near the cavity's outer walls.

Eight pure plutonium oxide samples, seven impure oxide samples, and eight plutonium metal samples were measured using the new multiplicity electronics package and the dual-mode counter in the high-efficiency mode. For the oxide samples, the predelay was set at 3 μs and the coincidence gate width was 32 μs. For the metal samples, the predelay was also 3 μs, but the gate width had to be decreased to 12 μs to keep the multiplicity circuit's sorting register from overflowing. All samples were centered in the sample cavity where the detector's response was most uniform. The oxide samples were measured for 5000 s each; the metal samples were measured for 1000 s each.

Because the multiplicities counted with the new circuit ranged from 1 to 32 for these samples and available deadtime correction schemes could not provide accurate correction for multiplicity distributions this large, a new scheme was derived so that measurements from the new circuit could be properly corrected. This new technique by Dytlewski is described in Ref. 4. All assays were based on measured multiplicity distributions corrected using this approach. The

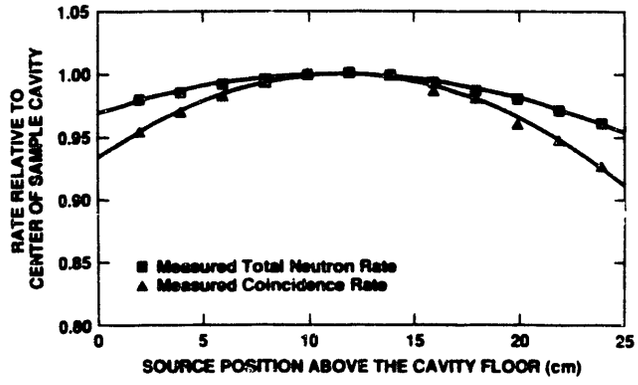

Fig. 2. Axial response to a ^{252}Cf source of the dual-mode multiplicity counter in the high-efficiency mode.

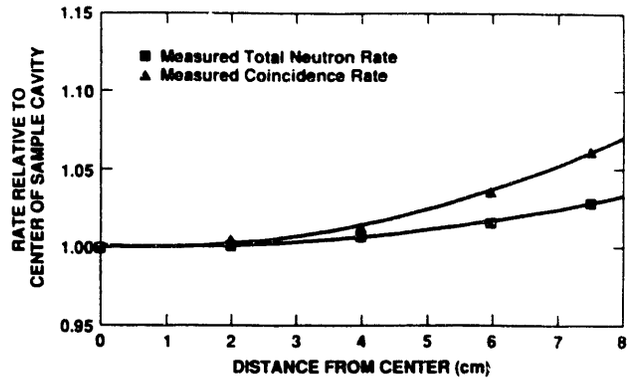

Fig. 3. Radial response to a ^{252}Cf source of the dual-mode multiplicity counter in the high-efficiency mode.

analysis of the corrected multiplicity distributions to yield assay results was based on the emitted moments calculated by Boehnel /5/ and Cifarelli and Hage /6/. The calibration parameters were the detector's efficiency for a nonmultiplying, pure plutonium oxide sample; gate fractions derived from the die-away response to ^{252}Cf; and known values for the first, second, and third induced-fission moments for ^{239}Pu at 2 MeV.

The multiplicity assay results for the 23 samples are given in Table I. Figures 4 and 5 give the assay results derived as above for these samples relative to their known reference values. Included in these plots, for comparison, are the assay results obtained using conventional passive, neutron coincidence assay methods. For the oxide samples, the conventional two-parameter assay analysis included a multiplication correction based on a value for α calculated from the known isotopic ratios for the samples. For the metal samples, no multiplication correction was performed. The calibration parameters for the conventional assays were derived from the characterization with ^{252}Cf and a relationship derived for the pure oxides between the multiplication-corrected coincidence rate and the known ^{240}Pu-effective mass.

Table II gives a comparison of the overall assay results obtained for these samples with the multiplicity approach to the conventional assay results. The improvement attained with the multiplicity analysis is striking. The results of the multiplicity assay compared to reference values for the pure and impure oxide samples display no statistically significant bias, and the scatter in the results is consistent with the expected uncertainty in the reference values and the statistical precision predicted by Ensslin et al. /3/ A more recent effort has been made to predict the statistical precision of multiplicity assays from measured quantities. The results of this effort are reported in Ref. 7.

The metal sample results display a bias and their scatter is slightly larger than expected. Although the reason for these discrepancies is not yet completely understood, several factors have been identified that may be responsible for them. One factor is that the reference values for three of the samples were suspect. These samples are starred in Table I. However, removing the results for these samples from the calculation of the average and standard deviation of the assays only improves the latter. The bias remains about the same.

Several factors may contribute to this bias. One factor is that the die-away response for this counter was not exponential. Figure 6 displays the measured die-away response for this counter for ^{252}Cf with an exponential fit overlayed. At early times the die-away is faster; at later times there is a

TABLE I. Multiplicity Assay Results Obtained with the Dual-Mode Multiplicity Counter in the High-Efficiency Mode

Sample Type	Total Pu (g)	^{240}Pu-effective (g)	M	Alpha	Multiplicity Assay/Reference
Pure oxide	59.97	10.10	1.025	0.436	0.993
Pure oxide	171.9	29.29	1.038	0.426	1.001
Pure oxide	321.9	54.33	1.050	0.418	1.009
Pure oxide	384.8	65.10	1.056	0.416	1.002
Pure oxide	543.8	92.14	1.070	0.416	1.010
Pure oxide	612.7	104.5	1.078	0.418	1.013
Pure oxide	848.6	144.3	1.097	0.420	1.006
Pure oxide	876.6	149.1	1.084	0.413	1.011
Impure oxide	19.97	1.977	1.014	0.807	1.000
Impure oxide	74.89	7.414	1.022	0.766	1.005
Impure oxide	149.8	14.83	1.031	0.766	0.999
Impure oxide	299.6	29.66	1.043	0.747	1.014
Impure oxide	458.4	43.33	1.057	0.705	1.005
Impure oxide	614.0	64.95	1.063	0.710	0.998
Impure oxide	777.7	81.31	1.074	0.649	1.014
Metal	585	34.46	1.208	0.858	0.836
Metal	1000	57.66	1.590	0.024	0.936
Metal	1352[a]	81.66	1.119	0.847	1.021
Metal	1678[a]	98.33	1.175	0.682	0.830
Metal	1729[a]	101.7	1.410	0.198	0.912
Metal	2000	115.3	2.070	0.113	0.845
Metal	2200	130.8	1.611	0.086	0.898
Metal	2962	141.3	2.086	0.112	0.873

[a]Reference values may not be accurate

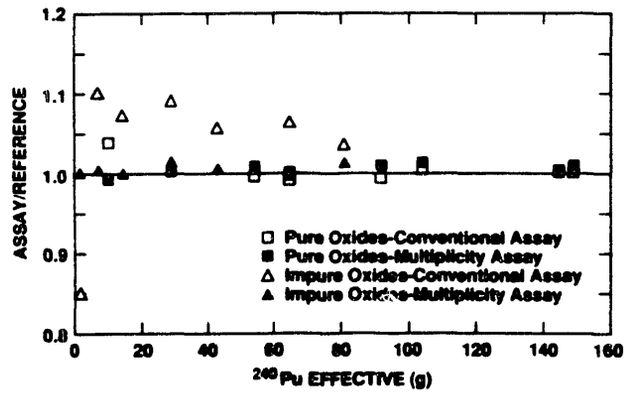

Fig. 4. A comparison of the multiplicity assay results obtained with the dual-mode multiplicity counter in the high-efficiency mode to conventional assay results for plutonium oxide samples.

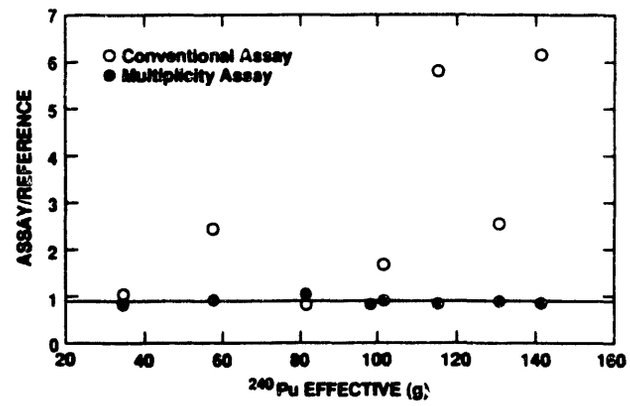

Fig. 5. A comparison of the multiplicity assay results obtained with the dual-mode multiplicity counter in the high-efficiency mode to conventional assay results for plutonium metal samples.

TABLE II. Comparison of the Overall Multiplicity Assay Results Obtained with the Dual-Mode Multiplicity Counter in the High-Efficiency Mode to Conventional Assay Results		
Average Assay/Reference with Standard Deviation		
Sample Type	Conventional Assay Results	Multiplicity Assay Results
Pure plutonium oxides	1.004 ± 1.4%	1.006 ± 0.66%
Impure plutonium oxides	1.039 ± 8.2%	1.005 ± 0.68%
Plutonium metal	2.665 ± 81%	0.894 ± 7.1%
Plutonium metal (subset) using relative calibration		0.975 ± 4.9%

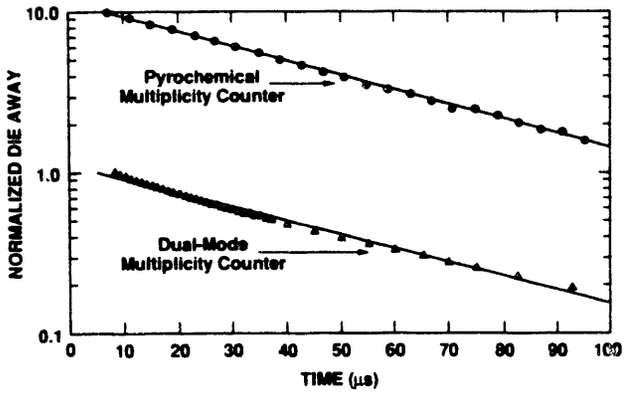

Fig. 6. The die-away response of the dual-mode multiplicity counter in the high-efficiency mode and the pyrochemical multiplicity counter for ^{252}Cf. The former is normalized to 1; the latter is normalized to 10.

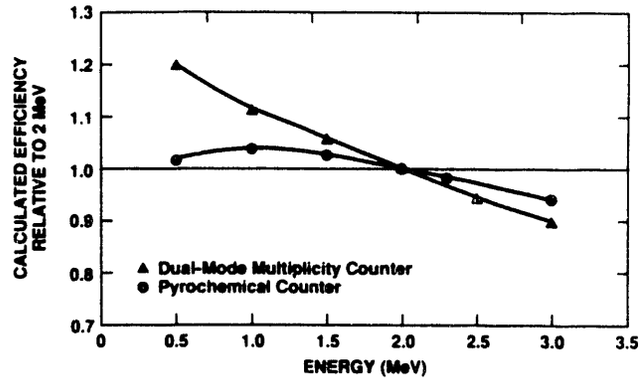

Fig. 7. The calculated efficiency of the dual-mode multiplicity counter in the high-efficiency mode and the pyrochemical multiplicity counter for a monoenergetic point neutron source in the center of the sample chamber.

tail. This behavior complicates the determination of the effective die-away time for the counter and the subsequent determination of the gate fractions needed to analyze the detected multiplicity distributions. The oxide assays are not as sensitive to errors that may result as a consequence of this complex die-away behavior because their third moment is small (that is, their self-multiplication is small). However, the metal assays are much more sensitive to such errors, because their third moments are substantially higher relative to their first and second moments.

Another possible source of the bias is that the average energy of the neutrons emitted by the metal samples is probably slightly different from that for the oxide samples. The multiplicity assay calibration was based on a small plutonium oxide source. Spontaneous-fission neutrons from ^{240}Pu have a mean energy of ~2.0 MeV. Induced-fission neutrons have a mean energy of ~2.2 MeV; (α,n) neutrons from oxygen have a mean energy of ~1.8 MeV. The average energy of the neutrons emitted by these samples will be a complex combination of these neutron energy spectra and spectra resulting from (α,n) reactions with any other impurities. The calculated energy response of the dual-mode counter in the high-efficiency mode relative to 2 MeV is given in Fig. 7. These calculations were made with MCNP version 3B /8/ for a monoenergetic point source placed in the center of the sample chamber. From these calculations, if the mean energy of the neutrons emitted by the metal samples is larger on average than for the oxide samples, the detection efficiency of the dual-mode counter will be lower and the assay will be biased low. A multiplicity analysis will be especially sensitive to such effects because the detected third moment depends on the third power of the detection efficiency.

The metal assays were re-evaluated, for the subset of samples whose reference values were believed to be accurate, using a relative calibration based on a small metal sample whose multiplication was known. Using this relative calibration approach instead of a calibration based on plutonium oxide substantially reduces the bias, which suggests that the bias is indeed caused by an effect related to the general sample composition. The overall results for this relative calibration method are included in Table II. More study is needed to understand the effect of sample composition on multiplicity assays.

3. The Pyrochemical Multiplicity Counter

The assay results discussed above suggest that not only does a multiplicity counter need to have a high efficiency and a low die-away time, it should also have a detection efficiency that does not vary with the emitted neutron energy spectrum and a well-behaved, exponential die-away response. The dual-mode counter was designed as a

research tool and was not optimized in this sense. Also, its sample cavity is too small to accommodate the large range of container types that may be encountered in facilities.

A second counter has been designed and built at Los Alamos to be more suited to in-plant use and to optimize, as much as reasonably possible, the parameters that have thus far been identified as important in multiplicity assays by thermal-neutron detectors. We determined the optimum arrangement of tubes in the body of the counter and the best choice for end-plug materials to yield a high and invariant detection efficiency by using Monte Carlo simulations. These calculations are reported in detail in Ref. 9. The body of the counter was constructed only from polyethylene to simplify its die-away response. In Figure 7 we compare the calculated energy response of this new counter to that of the dual-mode counter.

The new counter, named the pyrochemical multiplicity counter, consists of 126 ^{3}He tubes with 71-cm active lengths arranged in four rows. Figure 8 gives a schematic diagram of this new counter as it was conceived after the calculations. The cadmium-lined sample cavity is 24 cm in diameter and 38 cm high, and its efficiency profile was optimized for cans up to 20 cm wide and 36 cm high. The end plugs are made of graphite. To allow the counter to be used for both at-line and in-line applications, it was built in two identical halves. To accommodate the physical separation of the halves, one tube from the third ring was moved to the outermost ring and the tubes in each ring were moved together slightly to allow for the split in the junction box. The calculations were not repeated for this new configuration because we did not expect it to significantly affect the results. Each half in the final counter then contains 63 tubes. The halves can be brought together to form a single free-standing counter or can be split and then placed around a glove-box well for in-line applications.

The calculations predicted that the counter would have an average 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% for ^{252}Cf and its measured die-away time is 47.2 μs. The differences between the calculations and the measured quantities are consistent with the above-mentioned design changes. The measured die-away response of the pyrochemical multiplicity counter is given in Fig. 6 along with an exponential fit. These data do not display the nonexponential behavior of the dual-mode counter.

Figures 9 and 10 show the spatial response of the pyrochemical counter measured with a ^{252}Cf source centered in the sample chamber and moved axially and radially. Several of the calculated points are overlayed for comparison. This detector's response is more uniform in the central and lower regions of the cavity than the dual-mode counter and increases less near the cavity's outer walls. The agreement between the calculations and measurements suggests that the Monte Carlo model may also be useful to understand sample-related variations in the detector response that cannot practically be ascertained any other way.

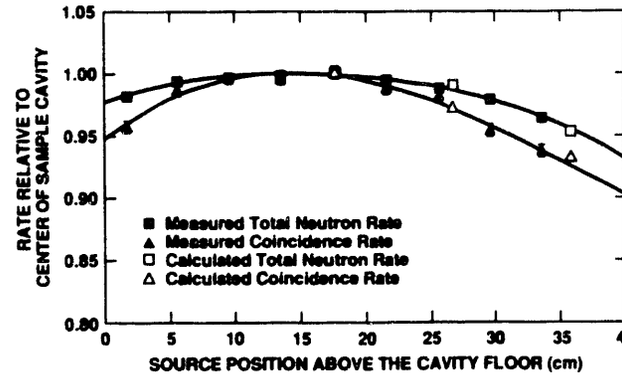

Fig. 9. Axial response of the pyrochemical multiplicity counter to a ^{252}Cf source.

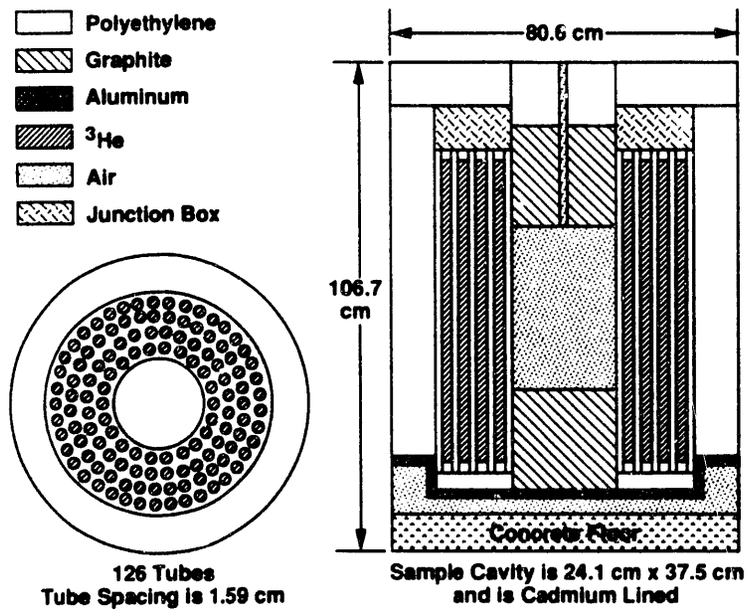

Fig. 8. Schematic of the pyrochemical multiplicity counter.

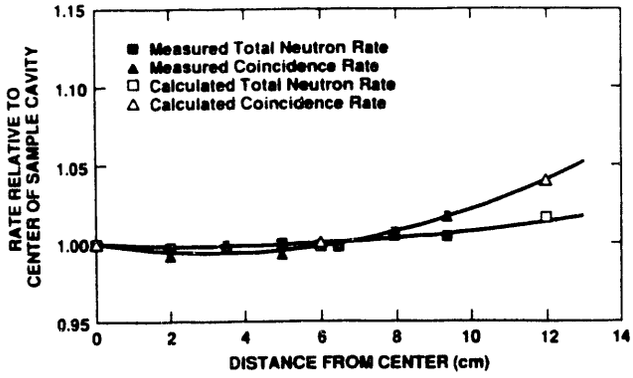

Fig. 10. Radial response of the pyrochemical multiplicity counter to a ^{252}Cf source.

4. Future Development

Although these early characterization studies demonstrate that the goals identified for the new counter from the lessons learned from the dual-mode counter have been achieved, actual sample measurements need to be performed in it. Some preliminary assays of very small samples have been successful, but measurements of more characteristic samples have been postponed until a new multiplicity electronics package is completed. This new package will allow for sorting multiplicities up to 256. Thus, larger coincidence gate widths can be used and the precision of the results for high-rate samples will improve. Also any effects as a result of fluctuations in the die-away response caused by the nature of the sample should be reduced by using longer gate widths.

Because it is probably not possible to build a thermal-neutron detector in which the detection efficiency does not vary, future measurements will include an analysis of correlations between sample size and composition and the relative counting rates of the individual rings of the pyrochemical counter. The effect of reducing the number of rings in a multiplicity counter will also be studied to determine if a multiplicity counter, which is smaller and less costly but perhaps is limited to a smaller range of sample types, is practical.

5. References

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