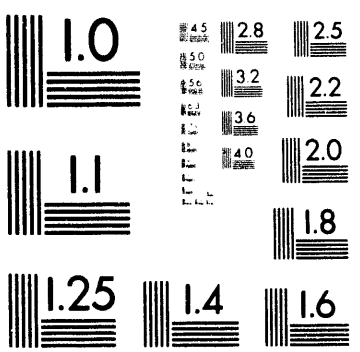
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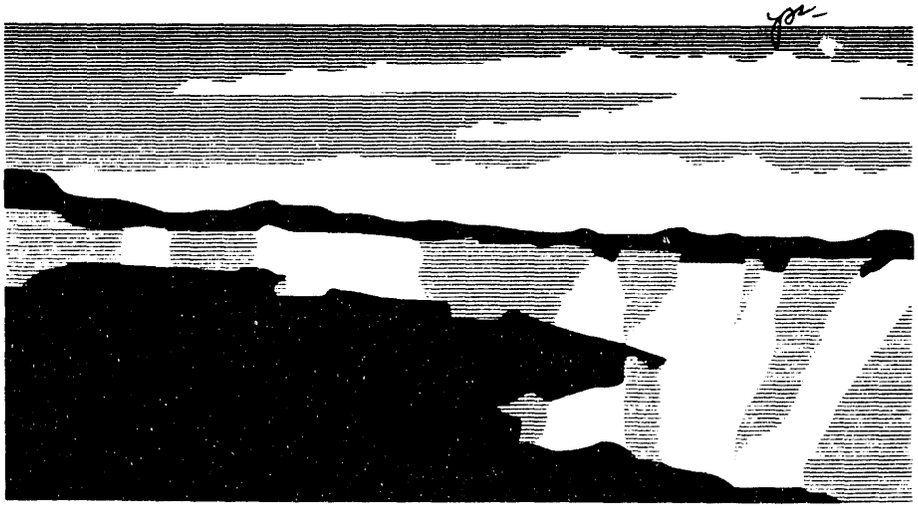

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THERMAL NEUTRON MULTIPLICITY MEASUREMENTS USING THE PYROCHEMICAL MULTIPLICITY COUNTER AT LAWRENCE LIVERMORE NATIONAL LABORATORY*

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ABSTRACT

The pyrochemical multiplicity counter designed and built at Los Alamos has been undergoing tests and evaluation at Lawrence Livermore National Laboratory (LLNL). Measurements have been performed using a variety of plutonium oxide and metal materials. The pyrochemical multiplicity counter uses the information contained in the higher moments of the neutron multiplicity distribution to deduce the three unknowns in the assay problem: ^{240}Pu-effective mass, (α,n) neutron rate, and self-multiplication. This is an improvement over conventional neutron coincidence counting, which must rely on some estimate of the (α,n) neutron rate or self-multiplication to deduce an assay result. Such conventional techniques are generally unsatisfactory for impure materials for which these quantities are unknown.

We present the assay results obtained with the pyrochemical multiplicity counter and discuss the procedures necessary to produce good assay results. Using these procedures, we have obtained assay accuracies of 1%–2% for oxide materials in 1/2 hour measurement times. We also compare these results to those that would have been obtained using conventional neutron assay techniques and discuss the correlations we have observed between assay results and the ratio of total neutron counts in the different rings of the pyrochemical counter.

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INTRODUCTION

The pyrochemical multiplicity counter is a high-efficiency, thermal neutron counter designed and built for in-plant evaluation of the multiplicity assay technique.[1-2] Together with its associated electronics and software, it can measure and analyze thermal neutron data in a conventional, two-parameter coincidence counting mode or it can use the higher moments of the neutron multiplicity distribution in a three-parameter analysis.

In the conventional mode, the counter system uses the neutron coincidence rate, the total neutron rate, and sample plutonium isotopics to deduce an assay. The rates are corrected for deadtime and neutron background. Then, a ratio of (α,n) neutrons to spontaneous fission neutrons is computed from the sample isotopics or some other knowledge of the sample. Finally, an assay value is calculated using Ensslin's multiplication correction equations[3] and calibration parameters based on measurements of pure plutonium oxide standards.

In the multiplicity mode, the counter system measures the distribution of multiplicities in the coincidence and accidental gates. These distributions are corrected for deadtime using a method developed by Dytlewski.[4] The moments of the multiplicity distribution are then computed using the equations of Cifarelli and Hage.[5] Finally, these moments are corrected for background, and an assay value is computed using Boehnel's point model equations.[6] In this mode, the ratio of (α,n) neutrons to spontaneous fission neutrons and the sample self-multiplication are calculated from the measurement data itself.

Sample isotopics are only used to deduce the total plutonium assay value from the measured ^{240}Pu-effective mass.

Calibration of the detector in the multiplicity mode consists of measuring its efficiency for pure plutonium oxide and measuring the coincidence gate fractions using ^{252}Cf. Californium is used to measure the gate fractions because in the pyrochemical counter the gate fractions are only very weak functions of neutron energy. However, plutonium is used to determine the detector's efficiency because efficiency varies more strongly with energy. Another reason that plutonium is used is because the plutonium will capture some of the fission neutrons and thus change the "effective" efficiency of the counter. For the pyrochemical multiplicity counter, which has been designed to be insensitive to variations in neutron energies, the latter reason is the most important.

The pyrochemical multiplicity counter underwent its first test and evaluation at the Los Alamos Plutonium Processing Facility in late 1991.[7] Twenty plutonium-bearing samples were measured. The multiplicity assays of oxide and metal materials were superior to conventional assays. The assays of five plutonium oxide samples agreed with reference values to within 1% (one sigma) for 3000-s measurement times. The assays of five plutonium metal samples agreed with reference values to within 5.1%, but these assays displayed a negative bias, which correlated with the sample self-multiplication. The assays of 10 molten salt extraction residues agreed with reference values to within 4.8%. These residues were not expected to assay as well using the multiplicity technique because of the high (α,n) neutron component of their total emitted neutron rates. Longer count times are necessary to improve the accuracy of multiplicity results for such samples.

In 1992, the pyrochemical multiplicity counter was sent to LLNL for further tests and evaluation. Here, we report on the results of assay measurements made of 59 samples.

MEASUREMENTS

The samples measured consisted of high and low burn-up, impure plutonium oxides and metals. The metal samples had a variety of geometries. We segregated the metal samples into two groups. Metal Set 1 consisted of molten salt extraction metal product buttons, which were broken into pieces before being placed in cans for measurement. Metal Set 2 consisted of buttons and metal castings that were not broken.

Each sample was measured twice for 900 s. Each 900-s assay consisted of 30 x 30-s measurement intervals, and each interval was subjected to a statistical test to reduce errors due to high-multiplicity cosmic ray events. The average precision due to counting statistics for these measurements was 0.5%–1%. In addition, the total neutron rate in each of the pyrochemical counter's four rings of ^{3}He tubes was measured for 30 s.

Assay results were calculated for each sample using conventional, two-parameter analysis and the three-parameter multiplicity analysis. For the oxide materials analyzed using the two-parameter analysis, the ratio of (α,n) neutrons to spontaneous fission neutrons was computed from the sample isotopics. For the metal samples, this ratio was assumed to be zero. Calibration parameters for the two-parameter analyses were based on measurements of pure, high burn-up plutonium oxide standards.

The assay results were then compared to reference values, which were obtained using calorimetry and gamma-ray isotopic measurements. These reference values are thought to be accurate to about 1%.

ASSAY RESULTS

Table I summarizes the assay results compared to reference values for the two-parameter analysis and the three-parameter multiplicity analysis. Figure 1 shows these results as a function of the ^{240}Pu-effective mass of the samples. The multiplicity assays are superior overall to the conventional two-parameter assays.

In the two-parameter assays, the effects of impurities in the samples are readily apparent. On average, the assays for the oxide samples are biased high due to impurities. Interestingly, however, the assays of the metal samples are biased low with the metals in Set 2 having a much larger bias than the metals in Set 1. The bias in the metals is caused by failure of the oxide-based calibration to accurately assay metals. If the metals are broken into pieces as in Set 1, however, the two-parameter assay provides better results than for the unbroken metal samples.

In the multiplicity assays of the oxide samples, the effects of impurities are not apparent, and the scatter in the assay results is only slightly larger than the uncertainties in the reference values plus counting precision. There is still a negative bias in the results for the metal samples. However, the bias in the metal results is smaller than for the two-parameter assays, and there is no difference in the

TABLE I. Comparison of Two-Parameter, Conventional Coincidence Assays with Three-Parameter, Multiplicity Assays

Sample Type	Number of Samples	(Assay - Reference)/Reference (%)			
		Average Results for Two-Parameter, Conventional Assay	1σ	Average Results for Three-Parameter, Multiplicity Assay	1σ
Low Burn-Up Metal	10	-18.0	15.2	-9.7	5.4
High Burn-Up Metal	4	-30.9	1.6	-8.1	0.9
Metal Set 1 (Broken)	5	-3.3	1.3	-9.0	5.6
Metal Set 2 (Unbroken)	9	-28.7	9.1	-9.4	4.2
All Metal Samples	14	-21.7	14.1	-9.3	4.6
Low Burn-Up Oxide	11	23.1	15.8	-1.2	2.5
High Burn-Up Oxide	34	5.6	6.4	0.3	2.0
All Oxide Samples	45	10.0	12.2	-0.1	2.2
All Samples	59	2.3	18.5	-2.2	4.9

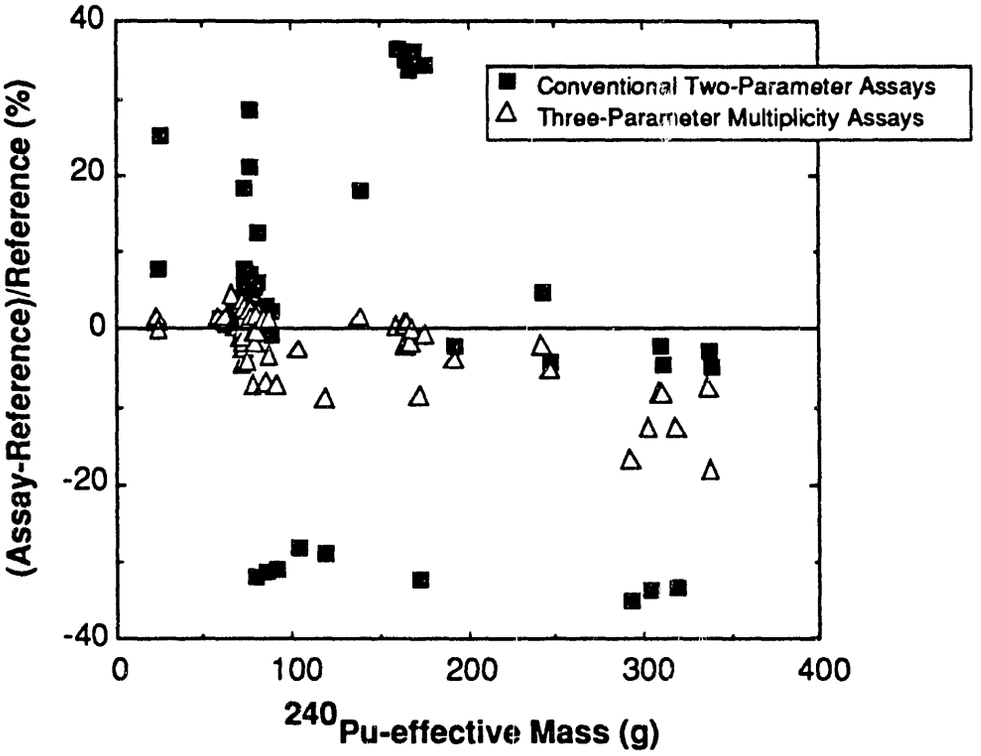

Fig. 1. Comparison of two-parameter conventional assay results to three-parameter multiplicity assay results from the pyrochemical multiplicity counter at Lawrence Livermore National Laboratory.

bias for the broken metal buttons compared to the metal samples that were not broken.

From Fig. 1 the negative bias in the results appears to correlate with sample mass. Figure 2 shows that, in particular, the assay bias is strongly correlated with the "measured" self-multiplication yielded by the point model equations. This is consistent with the behavior we observed in the earlier multiplicity measurements made with this counter.[7]

SELF-MULTIPLICATION DEPENDENT BIAS AND CORRECTION

We hypothesized that the multiplication dependent bias we observed in our three-parameter assay results is a consequence of an inadequacy of the point model to accurately describe neutron multiplication in large mass, dense samples. Specifically, the point model is based on the assumption that the probability of inducing fission for a neutron starting at any point in a sample can be approximated by a constant, global average probability. We believe this assumption breaks down for samples in which source neutrons have a mean free path that is shorter than or on the order of the average chord length through the sample.

To test this hypothesis, we modeled metal and oxide cylinders in the pyrochemical multiplicity counter using the Monte Carlo code, MCNP.[8] We performed calculations for a grid (Fig. 3) of points in each cylinder and obtained the calculated leakage multiplication, which results from starting spontaneous fission neutrons at each point on the grid. We then fit smooth curves to these multiplication values to obtain a multiplication "surface" as shown in Fig. 4. Finally, we integrated the point-model equations over the surface to obtain the total moments for the cylinders and used these total moments to predict an assay result. We found that for metal cylinders, the multiplication predicted by this procedure is higher than the true average leakage multiplication, and the assay result is biased low as a result. As the average

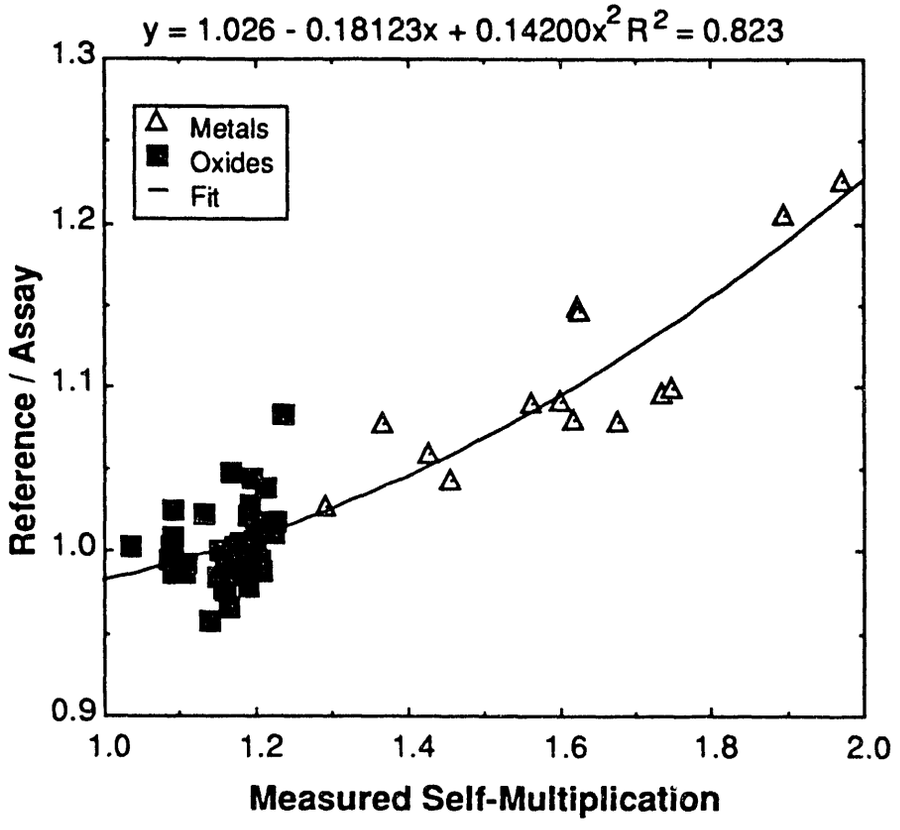

Fig. 2. Assay bias as a function of measured multiplication for three-parameter multiplicity assay results from the pyrochemical multiplicity counter.

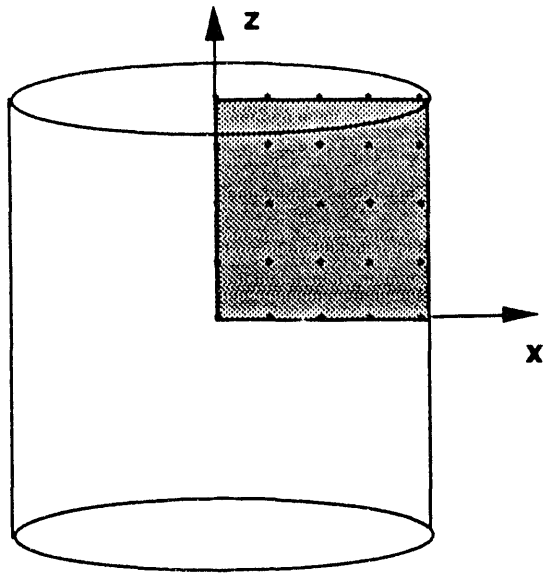

Fig. 3. Schematic of cylindrical sample geometry used to calculate leakage multiplication as a function of source neutron position.

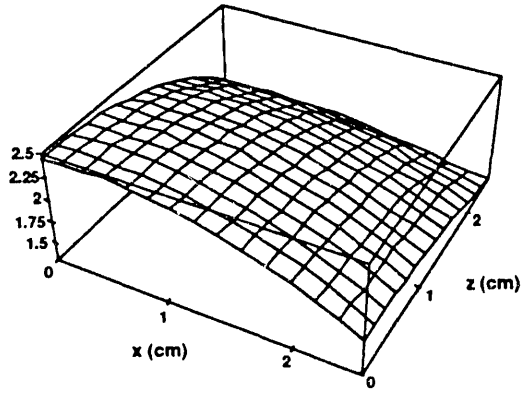

Fig. 4. Calculated leakage multiplication as a function of source neutron position for a 2-kg plutonium, right circular metal cylinder.

chord length through the metal sample increases, the bias increases. For the oxide cylinder, however, the multiplication obtained by integrating the surface is almost identical to the true average leakage multiplication. Thus, the point-model equations yield an accurate assay.

These results led us to the conclusion that a multiplication dependent correction to the multiplicity assays is necessary to achieve good assay results for samples with large plutonium masses. This correction is the quadratic fit shown in Fig. 2. We applied this correction to the point model assays. The corrected assay results are given in Table II and shown in Fig. 5. Now, the average bias in the assay results is zero, and the results for metals are only slightly poorer than for oxide samples.

RING RATIOS

To achieve high efficiency and neutron energy insensitivity, the pyrochemical multiplicity counter has four rings of ^{3}He tubes. The pulse stream from each ring is brought out of the counter separately so that the neutron rates in each ring may be counted. The innermost ring, Ring 1, is the most sensitive to low energy neutrons. The outermost ring, Ring 4, is the most sensitive to higher energy neutrons. We have concluded from calculations and experience with this counter that the ratio of the total neutron rates in these two rings is a sensitive indicator of variations in the mean energy of the neutrons emitted by a sample.[9] Thus, this ratio can be an indicator of sample-to-sample differences.

Figure 6 shows the multiplication-corrected, three-parameter assay results verses the ratio of the measured total neutron rates in these two rings. There is a grouping of the metal samples relative to the oxide samples with only a very slight overlap between the groups. This suggests that, on average, a metal sample can be distinguished from an oxide sample by its ring ratio. For these samples, only two have ring ratios that are close enough in value to make sample type classification on the basis of this ratio questionable.

There is also some correlation between the assay results and the ring ratios if the metal samples and the oxide samples are considered separately. However, the behavior of this correlation is not what one would predict on the basis of energy considerations alone. For each of these sample types, as the ring ratio increases the assay result decreases relative to reference values. From our calculations, if this ratio increases, the average energy of the emitted neutrons must be decreasing. The efficiency of the pyrochemical multiplicity counter increases slowly as neutron energy decreases. Thus, based on this argument, the assays should increase as a function of this ring ratio, not decrease. However, the decrease in the mean energy of the emitted neutrons caused by scattering is a function of the density of the sample and the average chord length through it. Neutron capture is also a function of these parameters. For these plutonium rich materials, as the average chord length and/or the density increases and neutron scattering increases, there is also more neutron capture by the plutonium. The net effect is to decrease the

TABLE II. Comparison of Corrected, Three-Parameter, Multiplictiy Assays					
		(Assay - Reference)/Reference (%)			
Sample Type	Number of Samples	Average Results When Multiplication Corrected	1σ	Average Results When Multiplicity and Ring Ratio Corrected	1σ
Low Burn-Up Metal	10	-1.4	1.9	-0.3	2.2
High Burn-Up Metal	4	3.1	0.8	0.9	0.9
Metal Set 1 (Broken)	5	-0.1	1.0	1.3	0.9
Metal Set 2 (Unbroken)	9	-0.2	3.3	-0.9	2.2
All Metal Samples	14	-0.1	2.7	0.0	2.0
Low Burn-Up Oxide	11	-1.6	1.9	-1.4	2.0
High Burn-Up Oxide	34	0.7	1.9	0.5	1.4
All Oxide Samples	45	0.1	2.1	0.0	1.8
All Samples	59	0.0	2.2	0.0	1.8

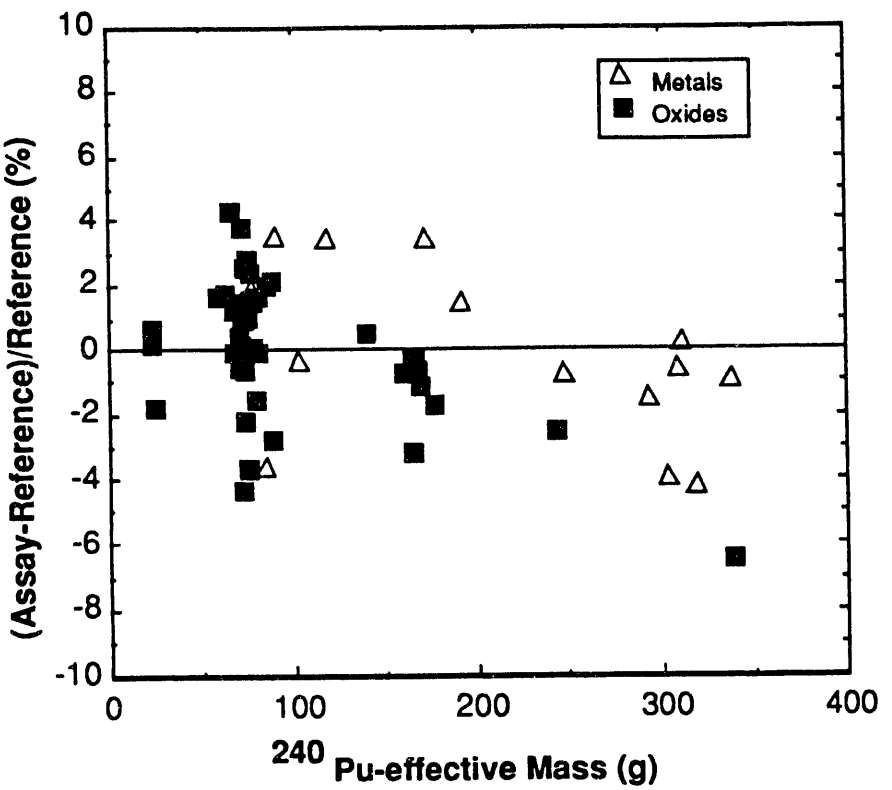

Fig. 5. Multiplication-corrected, three-parameter assay results from the pyrochemical multiplicity counter at Lawrence Livermore National Laboratory.

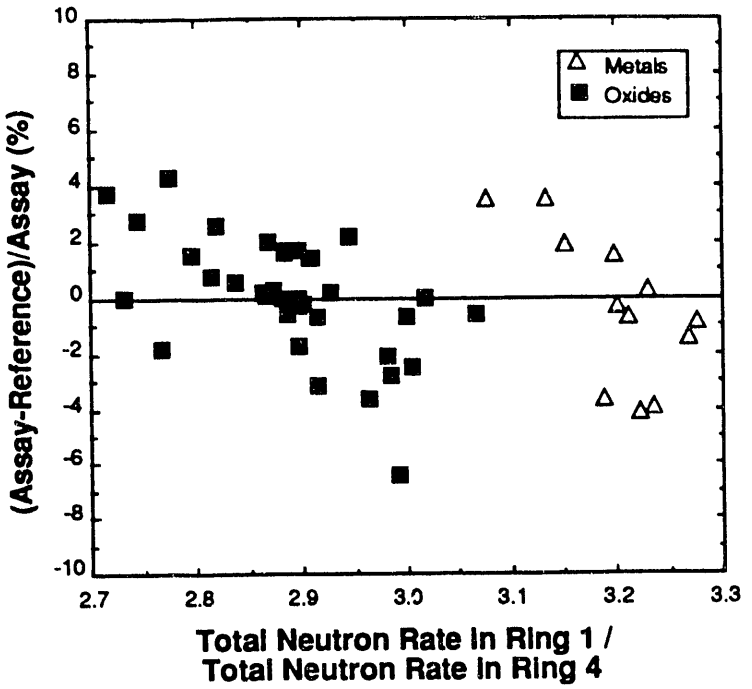

Fig. 6. The relationship between the ratio of the measured total neutron rate from the innermost ring to that for the outermost ring and multiplication-corrected, three-parameter assay results.

"effective" efficiency of the pyrochemical counter because of its relative insensitivity to changes in neutron energy. The assays results, thus, tend to decrease as the ring ratio increases as we see in Fig. 6.

If the ring ratio correlations are used to do one final correction to the assays, the improvement is small. These results are also given in Table II.

CONCLUSIONS

For impure plutonium bearing oxide samples, the pyrochemical multiplicity counter using three-parameter analysis provides assay results that are accurate and timely. Calibration of the detector using this technique does not require representative standards. For the 45 oxide samples measured during this evaluation, the pyrochemical multiplicity counter provided assays that were accurate to 2.2% (one sigma) in 1/2 hour measurement times.

The assays of large mass samples require multiplication dependent correction because of a limitation of the theory. This correction improves dramatically the assays of large mass samples. With this correction the pyrochemical multiplicity counter assayed all 59 samples to a one sigma accuracy of 2.2% in 1/2 hour.

Finally, the addition of ring ratio information to the assay analysis improves the assays slightly and helps to distinguish sample type. Of the 59 samples, 44 out of 45 oxide samples could be correctly identified as oxide by their ring ratio, and 13 out of 14 metal samples could be correctly identified as metal. With an additional ring ratio correction, the counter assayed all samples to a one sigma accuracy of 1.8%.

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