A New Neutron Multiplicity Counter for the Measurement of Impure Plutonium Metal at Westinghouse Savannah River Site

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A document prepared for INSTITUTE OF NUCLEAR MATERIALS MANAGEMENT at Naples, FL, USA from 7/26/98 - 7/30/98.

DOE Contract No. DE-AC09-96SR18500

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A New Neutron Multiplicity Counter for the Measurement of Impure Plutonium Metal at Westinghouse Savannah River Site

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Abstract

A new neutron multiplicity counter has been designed, fabricated, characterized, and installed for use in the assay of impure plutonium metal buttons from the FB-Line at the Westinghouse Savannah River Site (WSRS). This instrument incorporates the performance characteristics of the Pyrochemical or In-plant Multiplicity Counter with the package size of the Plutonium Scrap Multiplicity Counter. In addition, state-of-the-art features such as the de-randomizer circuit and separate ring outputs have been added. The counter consists of 113, 71 cm active length $^3$He tubes in a polyethylene moderator. Its efficiency for $^{252}$Cf is 57.8%, the highest of any multiplicity counter to date. Its die-away time is 50.4 ms and its deadtime is 50 ns. In this paper we will present the characterization data for the counter and the results of preliminary metal measurements at WSRS. We will also discuss the new challenges the impure metal buttons from FB-Line are presenting to the multiplicity counting technique.

Introduction

The FB-Line Neutron Multiplicity Counter (FBLNMC) evolved from multiplicity neutron detectors$^1,2,3$ developed at Los Alamos for the assay of impure plutonium samples. This new unit was designed to provide all state-of-the-art features in a single compact package. We designed the FBLNMC using the Monte Carlo Code for Neutron and Photon Transport (MCNP) to perform the Monte Carlo neutron calculations$^4$. The design goals for the FBLNMC were high efficiency (primary importance), uniform efficiency vs sample height, small die-away time, flat energy response, short dead time, and minimum overall size and weight. The first four of these design goals are generally in opposition to the last one. This instrument joins a suite of non-destructive assay equipment that are used for material control and accountability at FB-Line at Westinghouse Savannah River Site (WSRS).
Multiplicity Counter Design

Figure 1 shows a schematic diagram of the FBLNMC design with the one hundred thirteen $^3$He tubes surrounding the sample cavity with a diameter of 20 cm. The outer dimensions of the polyethylene (CH$_2$) shield are 66 by 66 by 80 cm. The total height is 92 cm. The sample cavity is lined with cadmium (0.8 mm thick) both to prevent thermal neutrons from returning from the CH$_2$ to the sample and to shield the $^3$He tubes from a possible high-intensity gamma-ray dose. There is no cadmium on the outside of the detector rings to reduce room-background levels. MCNP calculations$^5$ have shown that cadmium only reduces the totals background rate by $\sim$16%. However, the cadmium introduces its own background of coincident neutrons from cosmic-ray spallations and this is detrimental to assays of low mass samples. The end plugs shown in Fig. 1 are made of graphite to scatter the fast neutrons from the end zones back into the CH$_2$ detector volume. The MCNP calculation of the response of the detector system as a function of neutron energy is shown in Fig. 2 along with the comparison curve from the Pyrochemical Counter.$^2$ The majority of spontaneous fission and $(\alpha,n)$ reaction neutrons have energies in the range of 0.5-2 MeV. Certain impurities such as magnesium and beryllium produce (alpha,n) reaction neutrons that have higher energies. Assays of samples containing large quantities of these impurities may require additional corrections.

The 24 AMPTEK amplifiers that are used with the FBLNMC are shown in Fig. 3. Figure 3 also shows the cutouts for the four removable desiccant tubes that are used to keep the detector high-voltage junction box dry. To keep the rates in the four rings approximately equal and to minimize deadtime, fewer amplifiers service the outer rings than the inner rings.

The detector design includes two improvements over commercial multiplicity counters: a derandomizer circuit and an output from each individual ring of the detector. The former$^7$ reduces the deadtime of the counter by more than a factor of two. The latter provides input to two auxiliary scalars that can be used to diagnose sample anomalies$^8$. Figure 4 shows the relative rates as a function of energy that calculations predict each ring will detect. Ratios of the neutron rates in the rings provides a sensitive indication of the mean energy of the neutrons emitted by a sample and is strongly influenced by sample moderator or (alpha,n) reaction neutrons from many low atomic number impurities.

Calibration and Checkout

Before shipment to WSRS, the detector was characterized at Los Alamos using well-known $^{252}$Cf sources and plutonium oxide standards. The FBLNMC was calibrated for both multiplicity counting and the conventional coincidence counting "Known-Alpha" analysis$^9$ method. Table 1 gives the detector parameters that were derived from these measurements. Figure 5 shows assay results for 6 standards; 4 pure oxide samples and two impure samples. The conventional Known-Alpha assays were based on calibration
with pure oxide standards. As expected, the technique fails to give good results for the impure samples. Figure 6 shows the predicted performance of this counter for 30-minute count times and different material impurity levels as indicated by the ratio of (alpha,n) neutrons to spontaneous fission neutrons. The data in this graph were calculated using Ensslin" Figure-of-Merit Code\(^\text{10}\) for the detector parameters measured as part of the FBLNMC's characterization.

**Measurements at WSRS**

The FBLNMC was shipped to WSRS in the fall of 1997 and installed in FB-line. Since that time several plutonium standards have been measured to track its performance. New plutonium metal buttons and several sand, slag, and crucible samples have also been measured.

Figure 7 gives the results of multiplicity assay measurements of four plutonium oxide standards of varying purity over a 5 month period. From these data we conclude that the instrument is performing reliably and up to specifications.

Of the metal buttons, four have also been assayed using calorimetry to give reference values against which the multiplicity assays could be compared. This step is extremely important because of the bias problem that has been encountered with multiplicity assays of compact metals at other facilities\(^\text{11}\). The assay results for these four samples are given in Figure 8. The assays for these buttons include a bias correction based on the measured multiplication. A Known-Alpha analysis is included for comparison. This latter analysis assumed that the buttons were pure plutonium metal with zero (alpha,n) neutron emissions. Examination of these result reveals that the multiplicity bias correction was not adequate for the lowest mass sample. Figure 9 illustrates why. In this plot of the measured multiplication from the multiplicity analysis verses the assay bias observed in data taken over the last 8 years in three different instruments, the lowest mass Fl3-Line metal button displays an anomalous behavior relative to the other data. The reason for this is the unusually large (alpha,n) emissions from this sample. The multiplicity analysis suggests that the (alpha,n) emissions exceed the spontaneous fission emissions by about 25%. This compares to the other samples whose (alpha,n) emissions are no more than a few tens of percent of the spontaneous fission emissions. Calculations reported in reference 11 have predicted this behavior, but this is the first time it has been observed experimentally. Other new metal buttons measured at FB-Line have had (alpha, n) neutron emissions as high as 90% of their spontaneous fission emissions. So we plan to investigate this behavior further because we anticipate that a further bias correction that includes the (alpha,n) emissions will have to be implemented.

Six sand, slag, and crucible samples that had reference values from calorimetry were also assayed with the FBLNMC. These samples would not necessarily be considered good candidates for multiplicity counting because of their high (alpha,n) emissions. For these samples the ratio of (alpha,n) neutron emissions to that from spontaneous fission varied from 8 to nearly 30. Also, the mean energies of these (alpha,n) emissions is greatly
different than for plutonium metal or pure oxide. As a result the FBLNMC’s assays of these samples were biased by an average 7%. With the bias corrected, the assay results were as shown in Figure 10. These samples counted for an hour agreed on average to within 7% of the reference mass.

Conclusions

The FBLNMC has now been used to measure impure plutonium oxides, new FB-Line plutonium metal buttons, and sand, slag, and crucible samples. The instrument has performed to within specifications. Because of the impurities in the FB-Line buttons, an alpha dependence to the known bias behavior of the multiplicity assays has been discovered. We plan to investigate this further by obtaining reference values for a larger set of new buttons by calorimetry and studying the bias behavior in the FBLNMC’s assays.

Sand, slag, and crucible samples have also been studied. Although these samples have high (alpha,n) emissions, they assayed to within 7% of their reference masses in an hour count time.

Table

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<tr>
<th>TABLE I. FBLNMC Preliminary Calibration Parameters</th>
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<tr>
<td>Parameter</td>
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<tr>
<td>Efficiency for $^{235}$Cf</td>
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<tr>
<td>Efficiency for Pu</td>
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<tr>
<td>Die-away time (center)</td>
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<td>Predelay</td>
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<td>GateWidth</td>
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<td>High voltage</td>
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<td>Deadtime Coefficient a</td>
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<td>Deadtime Coefficient b</td>
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<td>Multiplicity Deadtime</td>
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<td>Doubles Gate Fraction</td>
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<td>Triples Gate Fraction</td>
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References


The sample cavity height is 4.1 cm and the Er-133 tubes and the graphite end plugs. The sample cavity is 20 cm x 41 cm and the diameter is 20 cm.

Figure 1: Schematic diagram of the BLNMC showing the location of the one hundred Er-133 tubes.
Fig. 2. MCNP calculations of the efficiency Vs the neutron energy for the FBLNMC and the Pyrochemical Counter.²
Fig. 3 Photograph of the high-voltage junction box including the AMPTEK boards.
Fig. 4. Relative Ring Responses of the FLLMCC.
Fig. 5. Assay comparison between conventional and multiplicity assays for FBLNMC.
Fig. 6. The expected precision for the FBLNMC for a 30 minute count time
Figure 7. Multiplicity assay results for plutonium oxide standards with the FBLMGC.
Figure 8. Assay results for 4 metal buttons with the FBLNMC.
Figure 9. Multiplicity assay results for metal buttons uncorrected for bias.
Figure 10. Multiplicity assay results for sand, slate, and crucible samples.