LA-UR- 95-3320

Title:

THE APPLICATION OF NEUTRON MULTIPLICITY COUNTING TO THE ASSAY OF BULK PLUTONIUM BEARING MATERIALS AT RFETS AND LLNL

Author(s):

D. G. Langner, M. S. Krick, and K. E. Kroncke

Submitted to:

American Nuclear Society
"Fifth International Conference on
Facility Operations—Safeguards Interface."
Jackson Hole, Wyoming USA
September 24-29, 1995
(FULL PAPER)

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

MASTER

Los Alamos
NATIONAL LABORATORY

Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the University of California for the U.S. Department of Energy under contract W-7405-ENG-36. By acceptance of this article, the publisher recognizes that the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. The Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy.

THE APPLICATION OF NEUTRON MULTIPLICITY COUNTING TO THE ASSAY OF BULK PLUTONIUM BEARING MATERIALS AT RFETS AND LLNL[a]

Diana G. Langner, Merlyn S. Krick, and Ken E. Kroncke
Los Alamos National Laboratory
Los Alamos, NM 87545 USA
(505) 667-2874

ABSTRACT

In the past several years, several facilities have identified a need for a large multiplicity counter to support safeguards of excess weapons materials and the measurement control and accountability of large, unusual samples. We have designed and fabricated two large thermal neutron multiplicity counters to meet this need at two DOE facilities. The first of these counters was built for Rocky Flats Environmental Test Site for use in the initial inventory inspection of excess weapons plutonium offered to International Atomic Energy Agency safeguards. The second counter was built for the Lawrence Livermore National Laboratory (LLNL) to support their material control and accountability program. For the LLNL version of the counter, a removable, fast-neutron interrogation assembly was added for the measurement of large uranium samples. In the passive mode these counters can accommodate samples in containers as large as a 30-gal. drum. This paper will report on the measured performance of these two counters and the data obtained with them.

I. INTRODUCTION

Prior to 1995, four neutron multiplicity counters (NMCs) had been designed and built at Los Alamos. Three are currently used for in-plant or international safeguards applications or both. The fourth counter is an experimental prototype and is used for training and research. The sample cavities of these counters can accommodate only relatively small samples. The largest item that can be measured optimally by the largest of these counters is 20 cm wide by 36 cm high.

In the fall of 1993, we began the design[1] of a prototype NMC to investigate the use of multiplicity counting for mass accounting of weapons components stored in ALR8 containers. This counter, the 30-gal.-Drum Neutron Multiplicity Counter, was scheduled to be completed in fiscal 1995. In July of 1994, Los Alamos was tasked by the Lawrence Livermore National Laboratory (LLNL) to build a similar counter for their material control and accountability program. Rather than procuring a custom-built counter, LLNL requested the 30-gal.-Drum NMC design to reduce cost. However, they asked that we add an active insert to the counter to research their need to measure uranium items.

In the fall of 1994, funding for the fabrication of the initial prototype 30-gal.-Drum NMC was terminated, and effort was redirected to aid Rocky Flats Environmental Test Site (RFETS) with their task to meet the Presidential Directive to offer excess weapons plutonium to international safeguards. The plutonium inventory offered by RFETS to IAEA safeguards consists of plutonium oxide of varying purities. The oxide is doubly contained. An 8801 type can that nominally contains 2 kg of plutonium in oxide form is doubly bagged and placed inside an 8802 can. The 8802 cans are stacked inside a steel tube or "spider" that self-centers in the 10-gal. drum. Usually there are two such cans to a drum. Because of the size of these containers, the 30-gal. NMC that was already undergoing fabrication was the only instrument that could be ready in time to perform the initial physical inventory verification (IPIV) then scheduled for March 1995.

The first 30-gal.-Drum NMC was delivered to RFETS in February 1995. Because of the short time available to complete and deliver this instrument, this counter could only be characterized using ^{252}Cf sources. The LLNL version of the 30-gal.-Drum NMC was more fully

---

[a]This work is supported by the US Department of Energy, Office of Nonproliferation and National Security, Office of Research and Development, On-Site Systems Branch.

characterized with both ^{252}Cf, uranium, and plutonium. It was delivered to LLNL in May.

The IPIV at RFETS has been delayed until December 1995. Installation and checkout of the LLNL counter was accomplished in September 1995. In this paper we will present the characterization data for these counters and discuss the performance of the LLNL version for the measurement of uranium and plutonium standards at LANL.

II. THE 30-GAL.-DRUM NEUTRON MULTIPLICITY COUNTER

The 30-gal.-Drum Neutron Multiplicity Counter is shown schematically in Fig. 1. A photograph of the LLNL version of the counter with its active insert is shown in Fig. 2. Each counter consists of one hundred twenty-six, 152-cm-active-length ^{3}He tubes embedded in a polyethylene moderator. Tube length, tube placement, and reflector materials were optimized using Monte Carlo simulations. Optimization criteria included a 40% detection efficiency relatively insensitive to sample-to-sample changes in the neutron energy of the emitted neutrons, a die-away time less than 60 μs, and a uniform response over the sample cavity.

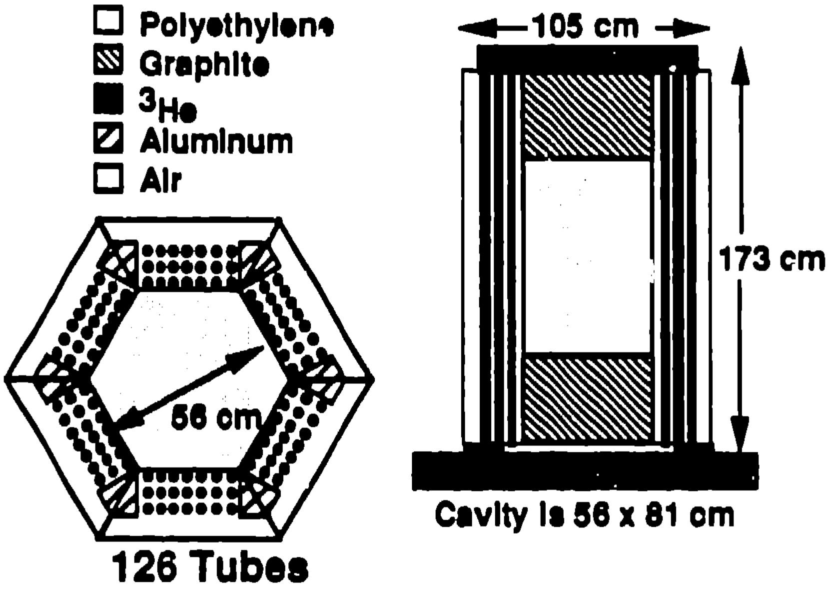

Fig. 1. Schematic diagram of the 30-gal.-Drum Neutron Multiplicity Counter.

Fig. 2. Photograph of LLNL's 30-gal.-Drum NMC showing the active insert.

Each sixth of the counter is a separate module. The counter modules are mated to each other via a large plate and hinge assembly that includes fork lift tine receivers. The lower graphite end plug of this counter is mounted on rails so that the end plug can be easily removed from the counter for sample loading. This end plug also has centering pins so that a 30-gal. drum can easily be centered in the sample cavity.

The active insert in the LLNL version of the counter is optimized for samples as large as 26 cm in diameter and 33 cm high. It consists of two cadmium-lined polyethylene cans each having a small, centered cylindrical cutout that will hold a standard Active Well Coincidence Counter

(AWCC)[2] AmLi neutron source. These cans are held in symmetric positions within the sample cavity by a framework that mates with the centering pins on the lower endplug.

The junction boxes for each module contain nine Amptek preamplifiers. The output signals from the inner, middle, and outer rows of tubes for each module are brought out separately. Then the signals from each module are input to a de-randomizing circuit[3] to produce the total signal from the counter. This total pulse stream is input into the multiplicity circuit of a Canberra 2150, and the signals from the inner and outer rows are input into its auxiliary scalars. With this arrangement, five independent quantities can be measured for each sample. Data are collected and analyzed using a new Windows neutron coincidence counting software package, NCCWIN, and a 486-DX2 IBM desktop computer.

III. PERFORMANCE CHARACTERISTICS FOR ^{252}CF

Both counters were initially characterized at LANL using a series of well-known ^{252}Cf sources. Detection efficiency, die-away time, deadtime, and spatial response were measured. The measured variations in the response of the counters relative to the center of the sample cavity are shown in Figs. 3 through 5. The responses for the horizontal dimension were measured along both the short axis (perpendicular to a module's face) and the long axis (from the cavity's center to a module's mating surface). These measurements agree well with predictions from Monte Carlo calculations. The measured detection efficiency, die-away time, and deadtime for each counter based on ^{252}Cf are given in Table I.

IV. PERFORMANCE CHARACTERISTICS FOR PLUTONIUM

In principle, a multiplicity counter can be completely characterized using measurements of well-known ^{252}Cf sources. In practice, however, we have observed from assays obtained with several multiplicity instruments that multiplicity assays based solely on ^{252}Cf data are biased low by 2-3%. We ascribe this bias to small uncertainties in the ^{252}Cf source strengths and small differences in the response of these thermal neutron counters to plutonium compared to californium.

It was not possible to measure plutonium standards in the RFETS counter prior to delivery. However, several Los Alamos standards were measured with the LLNL version. Two impure oxide standards and three pure standards were measured. Each of these samples was measured for several hours. In addition, these samples were measured in a variety of stacked configurations, and a single well-known metal disk was also measured. The stacked samples were measured for 30 minutes each and the metal was measured for an hour. Table II gives the multiplicity assay results for the single standard measurements using calibration

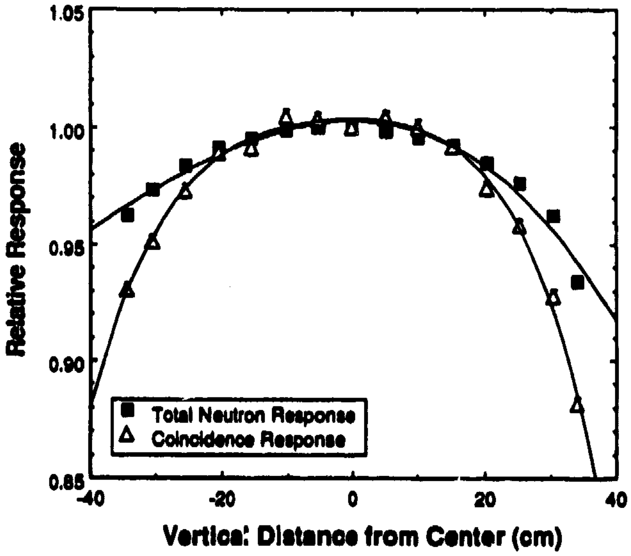

Fig. 3. Measured variations in the response of the 30-gal.-Drum NMC as a function of vertical position.

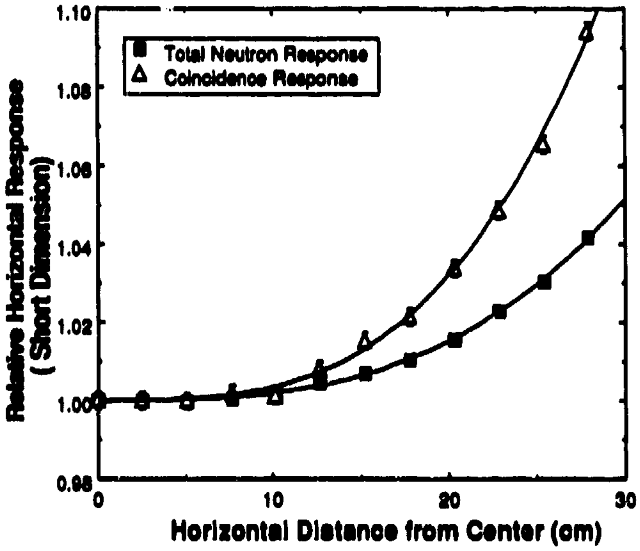

Fig. 4. Measured variations in the response of the 30-gal.-Drum NMC as a function of horizontal position in the dimension perpendicular to a module's interior face.

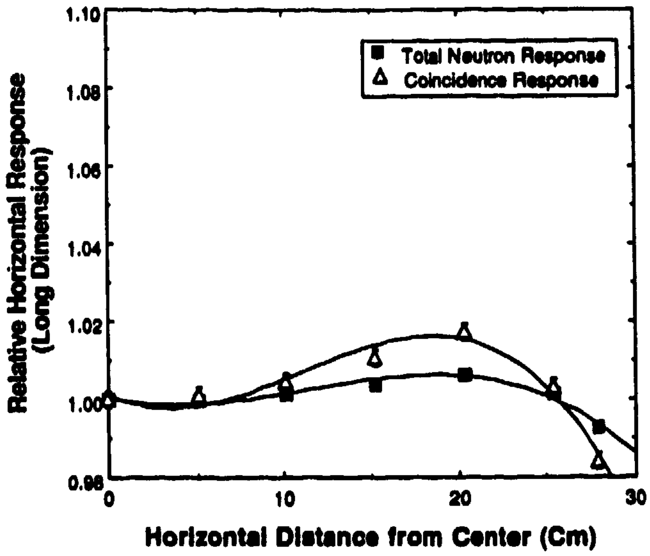

Fig. 5. Measured variations in the response of the 30-gal.-Drum NMC as a function of horizontal position in the dimension along a module's mating surface.

TABLE I. Characterization Parameters Based on ^{252}Cf		
Facility	RFETS	LLNL
Efficiency (%)	42.27	42.44
Die-away Time (ms)	55.4	54.9
Deadtime (ns)	25.0	24.5

parameters based only on ^{252}Cf. These data show the previously mentioned bias. Figure 6 gives the multiplicity assay results for all samples after adjusting the calibration by ~1.3% to plutonium. The five highest mass samples are the stacked oxides. Note that there is no significant bias in the assay results for the stacked samples relative to the single items. For comparison, assay results obtained using a conventional "Known-α" coincidence analysis are also plotted in this figure. Table III gives the calibration parameters for both instruments derived from these standard measurement results.

The ratio of the singles counts in the inner row to those for the outer row provides information about the mean energy of the neutrons emitted from the sample. This ratio decreases as the mean energy of the neutrons

TABLE II. Plutonium Standards Results for LLNL Counter at LANL		
^{240}Pu-eff (g)	Oxide type	Assay/ Reference
65.0	impure	0.963
81.5	impure	0.972
104.7	pure	0.974
144.6	pure	0.964
149.4	pure	0.975
Average		0.970
1σ		0.006

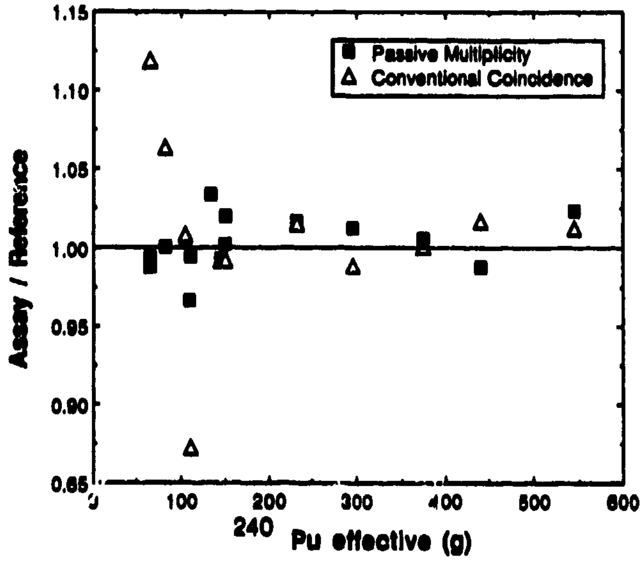

Fig. 6. Passive multiplicity and conventional "Known-α" assay results for the LLNL counter and plutonium standards measured at LANL.

TABLE III. Calibration Parameters for 30-gal.-Drum Neutron Multiplicity Counters		
Facility	RFETS	LLNL
Efficiency (%)	41.71	41.88
Doubles Gate Fraction	0.4100	0.4130
Triples Gate Fraction	0.1794	0.1820

increases. Figure 7 shows the passive multiplicity assays vs this ratio for the plutonium measurements. No correlations are evident in these data. This is consistent with the design criterion that the detection efficiency for this counter be relatively insensitive to sample-to-sample changes in the energy spectrum of emitted neutrons. However, there is a large difference between values of this ratio for the oxide samples relative to the metal sample. Also, the impure oxides have slightly lower values for this ratio compared to the pure samples. This is consistent with what has been observed with other instruments: that this ratio is a good indicator of sample differences.

V. ACTIVE MEASUREMENT OF URANIUM WITH THE LLNL COUNTER

Eight standard disks made of highly enriched uranium metal were measured in various stacked arrangements in the LLNL counter using the active insert loaded with two matched AWCC-type AmLi sources. Figure 8 shows the measured doubles rate vs ^{235}U mass for these measurements.

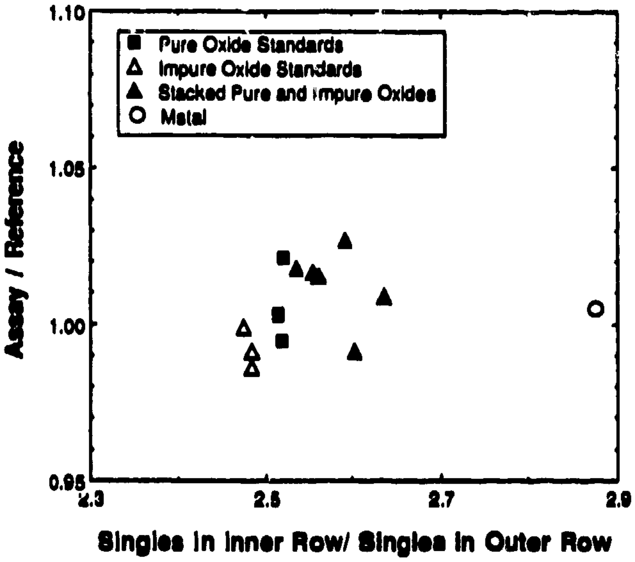

Fig. 7. The ratio of singles rates in the inner and outer rows of the LLNL counter for plutonium standards measured at LANL.

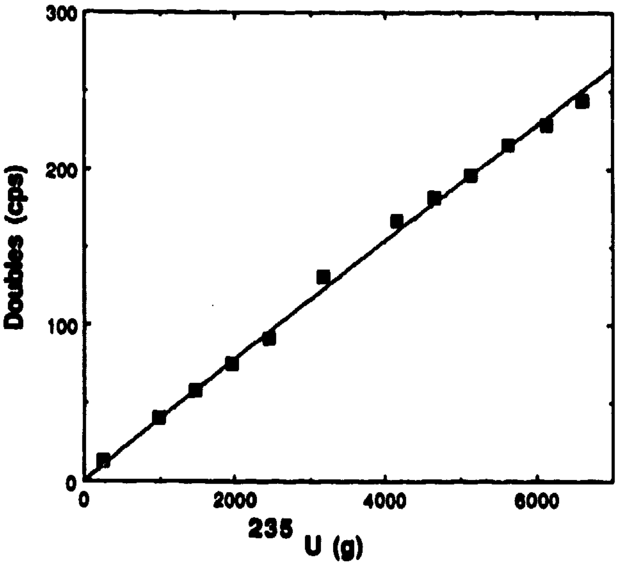

Fig. 8. Doubles rates for the LLNL counter vs ^{235}U mass for stacked highly enriched uranium metal disks and LANL AWCC sources.

These data are statistically linear. A quadratic polynomial was used to fit them, however, to account for the neutron self-absorption of the uranium as sample mass increases. This decision is barely statistically significant. Because of the open geometry of the insert relative to the large sample cavity of the 30-gal. counter and the relatively low mass range that these measurements represent, the usual cubic behavior of an active measurement is not observed in these data.

Using the method of Ensslin[4] and Krick,[5] neutron multiplication values for these disk stacks were computed from their triples to doubles ratios. Sample coupling was then computed from the doubles measurement corrected for multiplication. Figure 9 shows the relationship between these two quantities. This relationship is very similar to that reported by Krick for measurements of the same disksin an AWCC. This similarity is interesting because in active multiplicity counting, coupling and sample mass cannot be separated with an individual measurement. An empirical or calculated relationship must be used to reduce measured quantities to an assay.

To test the value of this relationship, the uranium disks were measured again using different stacking arrangements. Also, the AmLi sources were configured so that the source yield was different. The count times for these measurements were 15 minutes. The data were normalized to correct for the yield difference, the neutron multiplication was deduced as before, and the coupling was deduced from the multiplication using a fit to the data in Fig. 9. Finally, an assay value was computed using the multiplication-corrected doubles divided by the coupling. These assay results are plotted vs sample mass in Fig. 10. For comparison, assay results derived from the conventional doubles vs mass relationship are also plotted. The results from the two approaches are comparable. The conventional results have a 1 σ scatter of 3.3%. The active assays are only slightly worse with a scatter of 4%. Both techniques yield average assay divided by reference values that are statistically unity. The larger scatter in the active results is consistent with a loss in precision from using the triples information to deduce an assay.

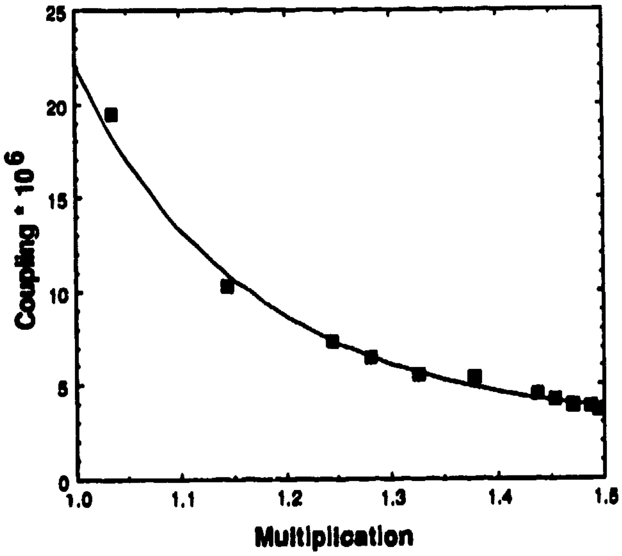

Fig. 9. Source coupling vs sample multiplication for stacked uranium metal disks in the LLNL counter.

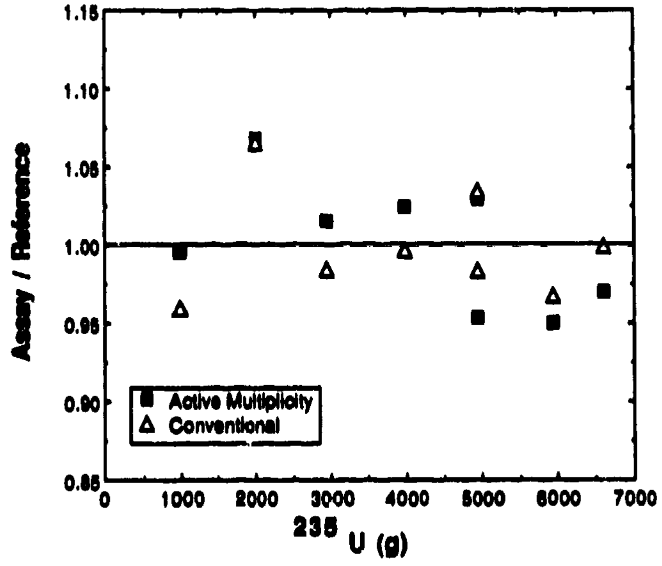

Fig. 10. Active multiplicity and conventional active assay results for stacked uranium metal disks in the LLNL counter.

VI. CONCLUSIONS

The 30-gal.-Drum NMCs fabricated for RFETS and LLNL meet or exceed design specifications. Their detection efficiencies and coincidence responses are very uniform throughout the sample cavity, and their assay performance is insensitive to sample-to-sample differences in the emitted neutron energy spectrum. In the passive mode, based on measurements of plutonium oxide standards, the instruments should produce assays of single oxide samples that are accurate to within a few percent. Up to 4 kg of plutonium, these results show no bias caused by sample stacking. As with previous instruments, the ratio of the inner- to outer-row singles counts provides information about the type of sample.

In the active mode, we have determined a preliminary calibration for the LLNL counter for uranium metal disks. The results using an active multiplicity interpretation of the data are consistent with the conventional active assays. Further investigation is necessary to determine whether the active multiplicity results are specific to sample type in this counter or are more generic.

ACKNOWLEDGMENTS

We would like to thank the staff of group NIS-6 at LANL for their support in the uranium and plutonium measurements made with the LLNL version of the 30-gal.-Drum NMC. We would also like to thank Mike Miller of group NIS-5 and his research associates, W. Feldman and R. Byrd of LANL group NIS-2, for their cooperation in sharing facilities and materials during these measurements.

REFERENCES

1. D. G. Langner, M. S. Krick, and K. E. Kroncke, "A Large Multiplicity Counter for the Measurement of Bulk Plutonium," *Nucl.- Mater. Manage.* **XXIII** (Proc. Issue), 474-479 (1994).

2. H. O. Menlove, "Description and Operation Manual for the Active Well Coincidence Counter," Los Alamos Scientific Laboratory report LA-7823-M (1979).

3. S. C. Bourret and M. S. Krick, "A Deadtime Reduction Circuit for Thermal Neutron Coincidence Counters with Amptek Preamplifiers, *Nucl.- Mater. Manage.* **XXIII** (Proc. Issue), 646-650 (1994).

4. N. Ensslin, M. S. Krick, W. C. Harker, M. C. Miller, R. D. McElroy, P. A. McClay, W. L. Belew, R. N. Ceo, L. L. Collins, Jr., and P. K. May,

"Analysis of Initial In-Plant Active Neutron Multiplicity Measurements," *Nucl. Mater. Manage.* **XXII** (Proc Issue), 465-470 (1993).

5. M. S. Krick, N. Ensslin, D. G. Langner, M. C. Miller, R. Sieblist, and J. E. Stewart, R. N. Ceo, P. K. May, and L. L. Collins, Jr., "Active Neutron Multiplicity Analysis and Monte Carlo Calculations," *Nucl.- Mater. Manage.* **XXIII** (Proc. Issue), 494-499 (1994).