Preprint
UCRL-JC-137317

Calibration of the Lawrence Livermore National Laboratory Passive-Active Neutron Drum Shuffler for Measurement of Highly Enriched Uranium Oxide

M.E. Mount, J. Glosup, C. Cochran, D. Dearborn
E. Endres

This article was submitted to
41st Annual Meeting of the Institute of Nuclear Materials Management
New Orleans, LA, July 16-20, 2000

U.S. Department of Energy

Lawrence
Livermore
National
Laboratory

June 16, 2000

Calibration of the Lawrence Livermore National Laboratory Passive-Active Neutron Drum Shuffler for Measurement of Highly Enriched Uranium Oxide

Mark E. Mount, Lawrence Livermore National Laboratory

7000 East Avenue L-347, Livermore, CA 94550 USA (925) 422-9800

Jeffrey Glosup, Lawrence Livermore National Laboratory

7000 East Avenue L-195, Livermore, CA 94550 USA (925) 423-0657

Curtis Cochran, Lawrence Livermore National Laboratory

7000 East Avenue L-347, Livermore, CA 94550 USA (925) 422-8542

David Dearborn, Lawrence Livermore National Laboratory

7000 East Avenue L-347, Livermore, CA 94550 USA (925) 424-6974

Eileen Endres, Lawrence Livermore National Laboratory

7000 East Avenue L-347, Livermore, CA 94550 USA (925) 423-6630

Abstract

In partial response to a Department of Energy (DOE) request to evaluate the state of measurements of special nuclear material, Lawrence Livermore National Laboratory (LLNL) evaluated and classified all highly enriched uranium (HEU) oxide items in its inventory. Because of a lack of traceable HEU standards, no items were deemed to fit the category of well measured. A subsequent DOE-HQ sponsored survey by New Brunswick Laboratory resulted in their preparation of certified reference material (CRM) 149 [Uranium (93% Enriched) Oxide - U_3O_8 Standard for Neutron Counting Measurements], a unit of which was delivered to LLNL in October of 1999. This paper describes the approach to calibration of the LLNL passive-active neutron drum (PAN) shuffler for measurement of poorly measured/unmeasured HEU oxide inventory. Included are discussions of (1) the calibration effort, including the development of the mass calibration curve; (2) the results from an axial and radial mapping of the detector response over a wide region of the PAN shuffler counting chamber; and (3) an error model for the total (systematic + random) uncertainty in the predicted mass that includes the uncertainties in calibration and sample position.

Background

The Measurement Assessment Project (MAP) was undertaken in June 1996 by the Department of Energy (DOE) to evaluate the state of special nuclear material (SNM) measurements across the DOE complex. Each site was required to classify SNM in different compositions as well measured, poorly measured/unmeasured, difficult to measure, or non-amenable to measurement. Because of a lack of traceable highly enriched uranium (HEU) oxide standards, Lawrence Livermore National Laboratory classified all HEU oxides as poorly measured. As a result of the MAP and a subsequent survey of

Work performed under the auspices of the US Department of Energy by the Lawrence Livermore National Laboratory under Contract W-7405-ENG-48.

complex-wide standards needs by New Brunswick Laboratory (NBL), certified reference material (CRM) 149 was prepared by NBL. One unit of CRM 149, consisting of a blank can plus six cans nominally containing 0.5 kg, 1.0 kg, 1.5 kg, 2.0 kg, 3.0 kg, and 4.0 kg of 93.2% enriched U_3O_8 was delivered to LLNL in October of 1999.

LLNL has two non-destructive assay instruments capable of measuring its HEU oxide inventory: the neutron multiplicity counter (NMC) and the passive-active neutron drum (PAN) shuffler. The NMC is fully tasked in the performance of plutonium measurements so the PAN shuffler is the instrument of choice for measuring LLNL's HEU oxides.

Calibration Plan

The LLNL CRM 149 Standard Unit

Table 1 summarizes the pertinent certified mass and estimated fill height information for the unit of CRM 149 used by LLNL in calibration of the PAN shuffler. All containers are of equivalent inside diameter (12.17 ± 0.05 cm), outside diameter (12.23 ± 0.05 cm), height (17.78 ± 0.08 cm), and bottom thickness (0.032 ± 0.004 cm)[1, 2].

Table 1. Pertinent certified mass and estimated fill height information for the LLNL unit of CRM 149 [1, 2]

LLNL Standard Nomenclature	U_3O_8 mass (g)	U mass (g)	^{235}U mass (g)	Estimated fill height (cm)[1]
CRM149-05	500.00	421.72	393.03	1.79
CRM149-10	999.80	843.28	785.90	3.58
CRM149-15	1499.93	1265.12	1179.04	5.37
CRM149-20	2000.23	1687.10	1572.30	7.16
CRM149-30	3000.07	2530.41	2358.23	10.75
CRM149-40	3999.37	3373.26	3143.74	14.33

1. A "tap density" of 2.4 ± 0.1 g/cm³ was provided with the NBL Certificate of Analysis. With a container inside diameter of 12.17 cm, the resultant mass per unit length is 279 g/cm.

In order to satisfy LLNL Plutonium Facility containment requirements, each standard container was sealed within a one gallon food can over-pack. The configuration of each standard container was such that its axial midplane and the food can were co-located and resting atop the inside surface of the bottom end cap, an estimated 0.36 cm above the food can base. Maintenance of this position is ensured through placement of aluminum foil between the standard container outer surface walls and lid and the food can inner surface walls and lid. Figure 1 shows a typical arrangement of the standard and one gallon food can containers.

Sensitivity of Measurement Results to Sample Axial and Radial Location within the Assay Chamber

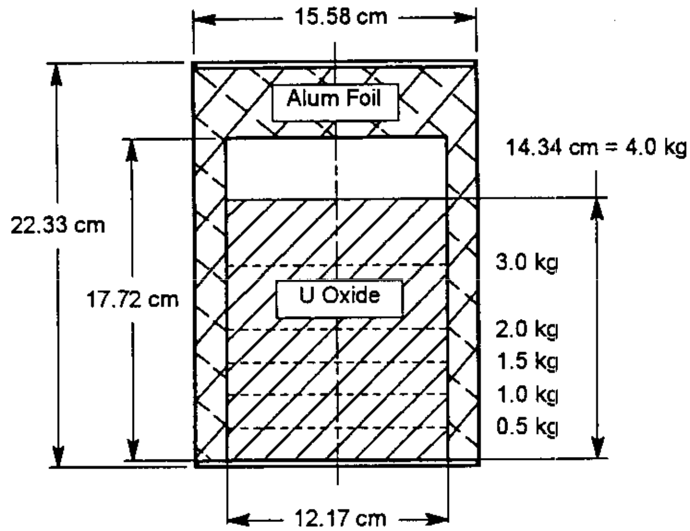

Figure 1. CRM 149 container and packaging

The sensitivity of measurement results with respect to the axial and radial location of an item within the assay chamber was examined by experiment. By corollary, a secondary outcome of this investigation was identification of the axial and radial region within the assay chamber where item location has no statistically discernible effect on measurement results.

Because the radial extent of source material in all standards is identical (6.08 cm) and the fill height of the CRM149-05 standard (1.79 cm) is the least of those in the CRM 149 unit, it was the standard of choice for the sensitivity studies. As such, it was measured at fixed locations over as much of the assay chamber axial height and radial extent as practical. An adjustable aluminum sample stand was used to fix the standard container at the appropriate height. For the radial sensitivity study, all measurements were performed with the sample stand platform located at 48.97 cm, the location where the radial and axial midplanes of the PAN shuffler assay chamber (chamber center) and each CRM 149 standard container are co-located.

A minimum of three PAN shuffler measurements were performed at each axial and radial location. A standard assay time was used for each measurement, consisting of a nominal 270 s background count followed by 34 shuffles (cycles of irradiation and counting), each with a 20.8 s period (a 1.4 s forward transfer time, an 11.7 s ^{252}Cf source irradiation time, a 0.8 s reverse transfer time, and a 7 s count time).

Tables 2 and 3 summarize the CRM149-05 measurement locations in the axial and radial sensitivity studies, respectively, the average decay-corrected delayed neutron count rate (counts/s) at each location, and the relative response at each location to the average delayed-neutron count rate (c/s) at 48.97 cm.

To the axial (4.52 cm to 93.42 cm) and radial (30.53 cm) extent of these sensitivity studies, results show the uncertainties in their average decay-corrected delayed neutron count rate (counts/s) due to sample location to be on the order of 6.7 and 4.8 percent or less, respectively.

LLNL has a wide variety of containers available for packagings. The largest is a five gallon lard can: 32.38 cm outside diameter and 38.73 cm height. If placed within the PAN shuffler assay chamber, the radial and axial extent of the lard can, as measured from the radial midplane and 48.97 cm elevation, are 16.19 cm and 87.70 cm, respectively. For a region of this extent, the predicted uncertainty in the average decay-corrected delayed neutron count rate (counts/s) due to sample location is on the order of 3.5 percent or less.

Table 2. Summary of the CRM149-05 measurement locations in the axial sensitivity study and the results at each location

Axial location (cm)[1]	Average count rate (counts/s)	Relative shuffler response[2]
93.42	517.50 ± 2.57	0.933 ± 1.32 %
84.53	549.94 ± 1.93	0.991 ± 1.27 %
75.64	568.36 ± 3.59	1.024 ± 1.38 %
66.75	573.86 ± 2.00	1.034 ± 1.27 %
57.86	557.86 ± 7.51	1.006 ± 1.82 %
48.97	554.80 ± 6.78	1.000
40.08	553.56 ± 2.84	0.998 ± 1.33 %
31.19	540.06 ± 8.95	0.973 ± 2.06 %
22.30	530.22 ± 3.89	0.956 ± 1.43 %
13.41	538.52 ± 3.49	0.971 ± 1.38 %
4.52	541.55 ± 4.29	0.976 ± 1.46 %

1. Height of the sample stand platform above the PAN shuffler turntable.
2. Relative to the average count rate (counts/s) at 48.97 cm, the location where the radial and axial midplanes of the PAN shuffler assay chamber and each CRM 149 standard container are co-located.

Table 3. Summary of the CRM149-05 measurement locations in the radial sensitivity study and the results at each location

Radial location (cm)[1]	Radial extent (cm)[2]	Average count rate (counts/s)	Relative shuffler response[3]
0.00	6.08	554.80 ± 6.78	1.000
12.22	6.13 - 18.30	558.60 ± 2.73	1.007 ± 1.32 %
24.43	18.35 - 30.52	581.51 ± 3.83	1.048 ± 1.39 %

1. Radial point on the sample stand platform where the axial centerline of sample container is placed.
2. Extent of the sample stand platform radius over which the standard mass lays.
3. Relative to the average count rate (counts/s) at 48.97 cm, the location where the radial and axial midplanes of the PAN shuffler assay chamber and each CRM 149 standard container are co-located.

Sensitivity in Measurement Results to Two Samples within the Assay Chamber

The sensitivity of measurement results with respect to the presence of two items within the assay chamber was also examined by experiment. Secondary and tertiary goals of this investigation were further confirmation of relative insensitivity of measurement results to sample location and the impact container size (volume to surface area) has on items with identical mass but different shape.

To examine as broad a ^{235}U mass range as possible, a number of container combinations were used for the two-item sensitivity study: CRM149-40 plus CRM149-15, CRM149-30 plus CRM149-10, CRM149-20 plus CRM149-10, CRM149-15 plus CRM149-05, and CRM149-10 plus CRM149-05. Each combination was measured with the sample stand platform at 48.97 cm and as follows:

- one atop the other - the container with the largest ^{235}U mass is on the bottom and the radial midplanes of the assay chamber and two containers are co-located, and
- side by side - the radial midplane of the assay chamber and the radial midplane at the point of contact of the side-by-side containers are co-located.

Two side-by-side combinations (CRM149-30 plus CRM149-10 and CRM149-10 plus CRM149-05) were also measured with the containers separated by a distance of 4.52 cm. A minimum of three PAN shuffler measurements were performed on each configuration using the previously described standard assay time for each measurement. Average decay-corrected delayed neutron count rate (counts/s) results for each configuration were compared to the sum of the average decay-corrected delayed neutron count rates (counts/s) for two single containers in each combination.

To evaluate the impact of container size (volume to surface area) on items with identical mass but different shape, average decay-corrected delayed neutron count rate (counts/s) results for a single container with a mass equivalent to each combination were compared to the sum of average decay-corrected delayed neutron count rates (counts/s) for the two single containers in each combination. The average decay-corrected delayed neutron count rate (counts/s) for the mass equivalent of the CRM149-40 plus CRM149-15 combination is from a Monte Carlo calculation for 5.5 kg of CRM 149 material in a container of equivalent diameter (12.17 cm) and extended height (22.78 cm)[3].

Tables 4 summarizes the CRM149-05 container combinations in the two-item sensitivity study, the average decay-corrected delayed neutron count rate (counts/s) for each two-container and single container configuration, and the relative response of each to the sum of the average decay-corrected delayed neutron count rates (counts/s) for the two single containers in each combination.

To the ^{235}U mass (1178.93 to 4322.78 g) extent of the two-item sensitivity study, there is virtually no difference between the average decay-corrected delayed neutron count rate (counts/s) results for each two-container configuration and the sum of average decay-corrected delayed neutron count rates (counts/s) for two single containers in each combination. As such, neutron interactions within one container appear to be independent of the presence of the other. Furthermore, as the axial (28.09 cm) and radial (14.48 cm) extent of the mass in the largest two-container combination (CRM149-40 plus CRM149-15) are within the five gallon lard can dimensional limits, the relative insensitivity of measurement results to sample location is further confirmed.

The average decay-corrected delayed neutron count rates (counts/s) for single containers with mass equivalent to a two-container combination are always less than the sum of average decay-corrected delayed neutron count rates (counts/s) for two single containers in each combination. Moreover, the difference increases as the total mass decreases. As expected, PAN shuffler measurements are sensitive to the container size (volume to surface area) in which the item is being measured. The current calibration and prediction of ^{235}U mass are based on CRM 149 size containers. Further studies will be required where and when different containers sizes are used.

Table 4. Summary of the CRM 149 container combinations used in the two-item sensitivity study and the results for each combination

LLNL Standard Nomenclature	Configuration	Average count rate (counts/s)	Relative shuffler response[1]
CRM149(40+15)	sum of singles	4854.36 ± 5.14	1.000
	side by side	4856.72 ± 13.0	1.000 ± 0.29 %
	one atop another	4829.81 ± 11.1	0.995 ± 0.25 %
CRM149-55	single	4607.02 ± 92.1[2]	0.949 ± 2.00 %
CRM149(30+10)	sum of singles	3648.64 ± 7.02	1.000
	side by side	3634.48 ± 1.46	0.996 ± 0.20 %
	one atop another	3619.67 ± 4.47	0.992 ± 0.23 %
	separated side by side[3]	3649.60 ± 5.74	1.000 ± 0.25 %
CRM149-40	single	3454.14 ± 4.83	0.947 ± 0.24 %
CRM149(20+10)	sum of singles	2823.76 ± 1.84	1.000
	side by side	2816.56 ± 5.63	0.997 ± 0.21 %
	one atop another	2805.87 ± 5.48	0.994 ± 0.21 %
CRM149-30	single	2650.43 ± 6.78	0.939 ± 0.26 %
CRM149(15+05)	sum of singles	1955.02 ± 7.09	1.000
	side by side	1961.04 ± 13.0	1.003 ± 0.76 %
	one atop another	1944.15 ± 3.86	0.994 ± 0.41 %
CRM149-20	single	1825.56 ± 0.25	0.934 ± 0.36 %
CRM149(10+05)	sum of singles	1553.01 ± 7.11	1.000
	side by side	1550.20 ± 7.75	0.998 ± 0.68 %
	one atop another	1541.44 ± 2.47	0.993 ± 0.49 %
	separated side by side[3]	1562.14 ± 5.09	1.006 ± 0.56 %
CRM149-15	single	1400.22 ± 1.75	0.902 ± 0.47 %

1. Relative to the sum of average count rates (counts/s) for the two single containers in the specified combination.
2. Monte Carlo calculated results for 5.5 kg of CRM 149 material in a container of equivalent diameter (12.17 cm) and extended height (22.78 cm).
3. Containers separated by 4.52 cm (2.26 cm each from the assay chamber radial midplane).

Mass Calibration

All mass calibration measurements were performed using the previously described standard assay time for each measurement and the sample stand platform height fixed at 48.97 cm. The mass calibration model was derived from the previously described measurement results for each CRM 149 container plus the side-by-side and one-atop-another configurations of the CRM149-40 plus CRM149-15 combination. The mass calibration curve that best fits this data is given by Equation (1) where r is the decay-corrected delayed neutron count rate (counts/s) and m is the ^{235}U mass (g).

$$r = 123.5777 + 1.1083\,m - 1.5077x10^{-8}\,m^{3} + 3.2751x10^{-12}\,m^{4} \qquad (1)$$

This mass calibration curve is shown with the measurement results in Figure 2. The residuals from this fitted model (difference between measurements and fitted values in counts/s) are shown in Figure 3. These residuals show some minor variations in mean over the range of sample masses which contribute a small systematic error to the measurements. Failure to include the third and fourth order terms in the polynomial in Equation (1) would substantially increase this systematic error.

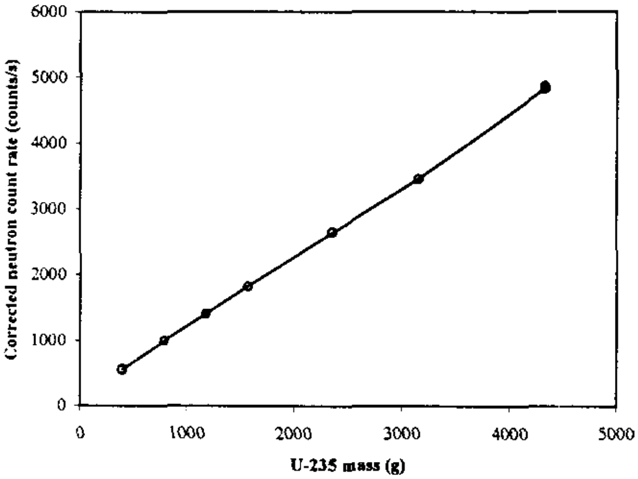

Figure 2. PAN shuffler uranium oxide mass calibration model versus ^{235}U mass

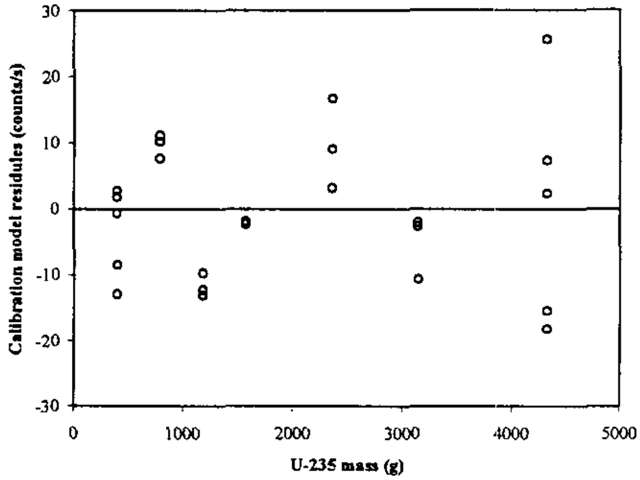

Figure 3. PAN shuffler uranium oxide mass calibration model residuals versus ^{235}U mass

The mass calibration curve in Equation (1) is used to estimate a ^{235}U mass from an observed count rate by inverting the equation. Presently, this inversion is performed using the root finding software built into Mathematica version 3.0. The inversion could also be accomplished numerically.

Error Modeling

The error in the measurement of the count rate can be divided into two basic types: a random uncertainty related to random fluctuations in the neutron counting and a systematic uncertainty caused by uncontrolled variations in the position of the nuclear material in containers and by minor inaccuracies in the calibration. By defining these uncertainties in terms of the decay-corrected delayed neutron count rate, they can be propagated through the calibration relationship to be expressed in terms of the ^{235}U mass.

The decay-corrected delayed neutron count rate r is derived from measurements of the raw delayed neutrons and the background. The algorithm used to calculate r is

$$r = (r_0 - b_0) f_{decay} f_{cycle} \tag{2}$$

where r_0 is the raw counting rate of delayed neutrons, b_0 is the background neutron counting rate, f_{decay} is the factor to account for decay of the ^{252}Cf source used to irradiate the sample, and f_{cycle} is the cycle correction factor used to adjust for the differences between nominal and actual irradiation and counting times. The random uncertainty in the counting rate arises from the random uncertainties in the quantities in Equation (2). The cycle correction and decay correction factors vary little and contribute only a negligible amount to the random uncertainty. The major contributor to random uncertainty is the counting statistics of the raw delayed neutrons and the background neutrons. The random uncertainty in r is propagated to provide a random uncertainty for the ^{235}U mass by

$$Var(m) = \frac{1}{(dr/dm)^2} \left(\frac{r}{r_0 - b_0} \right)^2 \left(\frac{r_0}{T_c} + \frac{b_0}{T_b} \right) \tag{3}$$

where T_c is the total counting time for the raw delayed neutrons, T_b is the background counting time and the counts are assumed to follow a Poisson distribution. For HEU oxides, the random uncertainty typically ranges from 0.2% to 1.2% of the ^{235}U mass.

The systematic uncertainty in the measurement can be subdivided into two pieces by the cause of the potential error: uncontrolled variations in placement of the nuclear material in the counting chamber and minor inaccuracies in the calibration relationship. Tables 2 and 3 indicate the size of potential errors in placement of nuclear material in radial and axial directions. However, the uncertainty added to the decay-corrected delayed neutron count rate by these potential errors is essentially negligible. Figure 3 shows minor inaccuracies in the calibration model that also contribute to the systematic uncertainty. These variations may be modeled as an additive bias in the calibration relationship. However, the size of this bias for any particular sample is not known. In a worst case, this bias behaves as a uniformly distributed random variable that is subtracted from the decay-corrected delayed neutron count rate before inverting the calibration relationship. Thus the systematic uncertainty becomes an additional variance term in the last term of Equation (3). The standard error for this random variable as estimated from the data is about 6 count/s. When this systematic uncertainty in the count rate is propagated to the ^{235}U mass, the total uncertainty in mass can range from 0.2% to 5% depending on the mass of the sample, with higher uncertainties at the lower masses.

References

[1] New Brunswick Laboratory, "Certified Reference Material Certificate of Analysis, CRM 149, Uranium (93% Enriched) Oxide - U_3O_8 Standard for Neutron Counting Measurements," New Brunswick Laboratory, Argonne, IL (November 30, 1999).

[2] Neuhoff, J., Personal communication, New Brunswick Laboratory, Argonne, IL (March 22, 2000).

[3] Rinard, P., Personal communication, Los Alamos National Laboratory, Los Alamos, NM (June 7, 2000)