NOTICE: This document contains information of a preliminary nature and is not intended for release. It is subject to revision or correction and therefore does not represent a final report.

ORNL/TM-2015/130

Direct Multiplication Measurement as an Alternative to Neutron Multiplicity Analysis

Robert D. McElroy, Jr.
Stephen Croft
Steve Cleveland

April 2015

Approved for public release; distribution is unlimited

OAK RIDGE NATIONAL LABORATORY
MANAGED BY UT-BATTELLE FOR THE US DEPARTMENT OF ENERGY

DOCUMENT AVAILABILITY

Reports produced after January 1, 1996, are generally available free via US Department of Energy (DOE) SciTech Connect.

Website http://www.osti.gov/scitech/

Reports produced before January 1, 1996, may be purchased by members of the public from the following source:

National Technical Information Service
5285 Port Royal Road
Springfield, VA 22161
Telephone 703-605-6000 (1-800-553-6847)
TDD 703-487-4639
Fax 703-605-6900
E-mail info@ntis.gov
Website http://www.ntis.gov/help/ordermethods.aspx

Reports are available to DOE employees, DOE contractors, Energy Technology Data Exchange representatives, and International Nuclear Information System representatives from the following source:

Office of Scientific and Technical Information
PO Box 62
Oak Ridge, TN 37831
Telephone 865-576-8401
Fax 865-576-5728
E-mail reports@osti.gov
Website http://www.osti.gov/contact.html

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

ORNL/TM-2015/130

Nuclear Security and Isotope Technology Division

DIRECT MULTIPLICATION MEASUREMENT AS AN ALTERNATIVE TO NEUTRON MULTIPLICITY ANALYSIS

Robert D. McElroy, Jr.
Stephen Croft
Steve Cleveland

Date Published: April 2015

Prepared by
OAK RIDGE NATIONAL LABORATORY
Oak Ridge, TN 37831-6283
managed by
UT-BATTELLE, LLC
for the
US DEPARTMENT OF ENERGY
under contract DE-AC05-00OR22725

CONTENTS

Page

LIST OF FIGURES v
LIST OF TABLES viii
ABBREVIATED TERMS x
ACKNOWLEDGMENTS xii
EXECUTIVE SUMMARY xiii
1. INTRODUCTION 1
2. ANALYSIS APPROACH 1
3. MEASUREMENT APPROACH 2
4. EARLY ATTEMPT AT THE DMM TECHNIQUE 5
5. EXPERIMENTAL CONFIRMATION (CURRENT STUDIES) 8
5.1 ^{252}Cf Shuffler Measurements 8
5.2 Large Volume Active Well Coincidence Counter 8
6. MEASUREMENT PRECISION FOR MOX ASSAY USING THE DMM TECHNIQUE 11
7. ACCURACY OF THE DMM TECHNIQUE FOR MOX ASSAY 12
7.1 BASELINE MCNPX MODEL 12
7.2 TWO ENERGY POINT MODEL 13
7.2.1 Determination of the Parameters **aε** and **ap** 16
7.3 IMPACT OF THE U:PU RATIO ON THE MULTIPLICATION MEASUREMENT 17
7.4 High Alpha and High U:Pu Ratio 19
7.5 DMM ACCURACY 21
8. CONCLUSIONS 21

LIST OF FIGURES

Figure **Page**

Fig. 1. Illustration of the source location for the ^{252}Cf source used for the Direct Measurement of Multiplication measurements. 3
Fig. 2. Plot showing the expected relationship between the sample self-multiplication as a function of the relative ^{252}Cf rates for the Direct Measurement of Multiplication assay with a PSMC. 4
Fig. 3. Sketch showing the measurement geometry of the early trials using a plutonium scrap multiplicity counter with mixed oxide samples. 6
Fig. 4. Plot of the scaled measured multiplication ,M, (determined from multiplicity analysis) as a function of ^{252}Cf source perturbation. 6
Fig. 5. Comparison of the Direct Measurement of Multiplication and traditional multiplicity analysis results using the plutonium scrap multiplicity counter. 7
Fig. 6. Plot showing the scatter in the observed count rate from the shuffler due to jitter in the source position. 8
Fig. 7. Screen shot of the MCNPX input file for the Large Volume Active Well Coincidence Counter (LAWCC) configured for the Direct Measurement of Multiplication test run (left) and a photograph of the LAWCC (right). 9
Fig. 8. Measured Totals neutron count rate and Reals rate ratios for the New Brunswick Laboratory U_3O_8 cans measured in the modified Active Well Coincidence Counter as a function of ^{235}U mass. 10
Fig. 9. Measured total and Reals rate ratios for the New Brunswick Laboratory U_3O_8 cans (listed in Table 1) measured in the modified Active Well Coincidence Counter as a function of the MCNPX-calculated self-multiplication for each standard. 10
Fig. 10. Expected measurement precision in the reported multiplication for multiplicity analysis using the ENMC (63% neutron detection efficiency) and for the Add-A-Source style multiplication measurement using a standard HLNCC-II (18% efficiency) and a 1×10^6 n/s ^{252}Cf interrogation source. 11
Fig. 11. Expected measurement precision in the reported plutonium mass for multiplicity analysis using the ENMC (63% neutron detection efficiency) and for the Add-A-Source style multiplication measurement using a standard HLNC (18% efficiency). 12
Fig. 12. Screen shot of the MCNPX input file used for the bias and precision studies (left) and a sectioned drawing of the HLNCC (right). 13
Fig. 13. Expected measurement performance of the Direct Measurement of Multiplication technique integrated with the standard point model (α = 1, 3, and 10 and U:Pu=1:1). 14
Fig. 14. Expected measurement performance of the Direct Measurement of Multiplication technique integrated with the two energy point model (α = 1, 3 and 10; U:Pu=1:1). 15
Fig. 15. Expected measurement performance of the Direct Measurement of Multiplication technique integrated with the two energy point model (α = 1, 3 and 10; U:Pu=1:1) using the Source 4C energy (α, n) distribution. 16
Fig. 16. Neutron detection efficiency as a function of energy for the HLNCC-II for a mono-energetic point source in the center of the HLNCC-II assay cavity. Overlain on the graph are the detection efficiencies for point sources of ^{240}Pu, ^{252}Cf and an oxygen (α, n) source. The measured efficiency for a ^{252}Cf point source [15] is plotted for comparison. 16
Fig. 17. Bias resulting from the Direct Measurement of Multiplication (DMM)/two-energy point model analysis integrated with a modified two energy point model for mixed oxide of various U:Pu ratios and with a fixed α (α =0.91). 17

Fig. 18. The relationship between the sample multiplication and the active interrogation ratio for a series of mixed oxide masses modeled at three different U:Pu ratios. 17
Fig. 19. The relationship between the sample multiplication and the active interrogation ratio for a series of mixed oxide masses modeled at three different U:Pu ratios. 18
Fig. 20. Expected measurement performance of the Direct Measurement of Multiplication technique integrated with a modified Two Energy Point Model [5] for MOX with ($\alpha = 0.91$) for various U:Pu ratios. 19
Fig. 21. The relationship between the sample multiplication and the active interrogation ratio for a series of mixed oxide masses modeled at three different U:Pu ratios for high alpha items. 19
Fig. 22. Expected measurement performance of the Direct Measurement of Multiplication technique integrated with the traditional point model and without the U:PU ratio correction for MOX items with ($\alpha = 10$) for various U:Pu ratios. 20
Fig. 23. Expected measurement performance of the Direct Measurement of Multiplication technique integrated with a modified Two Energy Point Model [5] for MOX with ($\alpha = 10$) for various U:Pu ratios. 20

LIST OF TABLES

Table **Page**

Table 1. Uranium standards from National Bureau of Standards and New Brunswick Laboratory (NBL) measured in the LAWCC 9

ABBREVIATED TERMS

AAS	Add-A-Source
AWCC	Active Well Coincidence Counter
DMM	Direct Measurement of Multiplication
ENMC	Epi-Thermal Neutron Multiplicity Counter
HEU	highly enriched uranium
HNLCC	High Level Neutron Coincidence Counter
LAWCC	large Volume Active Well Coincidence Counter
MCNP	Monte Carlo N-Particle software package
MOX	mixed oxide
NBL	New Brunswick Laboratory
NBS	National Bureau of Standards
NDA	nondestructive assay
ORNL	Oak Ridge National Laboratory
PSMC	plutonium scrap multiplicity counter
Pu	plutonium

ACKNOWLEDGMENTS

The authors would like to acknowledge and thank NA-241's Next Generation Safeguards Initiative (NGSI) within the Office of Nonproliferation and Arms Control (NPAC), National Nuclear Security Administration (NNSA), United States Department of Energy (DOE) for providing funding for this work.

EXECUTIVE SUMMARY

This report details the status and results of an investigation of Direct Measurement of Multiplication (DMM) as an alternative to neutron multiplicity counting of mixed oxide (MOX) materials. The study combines MCNP and direct measurement data to examine the feasibility and performance of the DMM technique. This work was performed under the Next Generation Safeguards Initiative DMM project.

Currently, the safeguards assay of plutonium and MOX scrap materials relies on the use of high-performance neutron multiplicity coincidence counting systems, each of which requires several hundred liters of ^{3}He with costs nearing $2 million. Both the limited ^{3}He supply and the high capital equipment cost of the multiplicity assay systems (dominated by ^{3}He costs) preclude the use of these once pervasive assay systems, resulting in a significant gap in our ability to safeguard MOX processing facilities. The primary objective of this project was to develop an alternative assay methodology to eliminate the need for high-performance multiplicity counting systems for the safeguards assay of MOX scrap.

Multiplicity analysis relies on counting single neutron (total) rates, and double and triple neutron coincidence rates to solve the point model equations for the three primary sample characteristics: mass, multiplication, and alpha (α), where α is the ratio of uncorrelated to correlated neutron emissions. It is the need to count three neutron coincidence rates that drives the need for very high-performance assay systems. These multiplicity coincidence systems—requiring very high neutron detection efficiencies (>60%) and short characteristic die-away times (<20 μs)—are the most challenging neutron assay systems to replace with alternative neutron detector technologies (indeed, to date, no suitable alternative detector technology has been identified). Rather than attempt to develop a replacement neutron detector matching the performance of ^{3}He, we have instead pursued an alternative measurement methodology using existing safeguards equipment.

The DMM technique relies on a point model analysis similar to traditional multiplicity assay; however, the multiplication of the item is determined by a secondary measurement, thereby obviating the need for measurement of three neutron coincidence rates and thus eliminating the need for a high-performance neutron multiplicity system. This work demonstrates that the necessary measurement performance can be achieved using a simple neutron coincidence system, such as the High Level Neutron Coincidence Counter (HLNCC) II, requiring <20 liters of ^{3}He rather than the 500 liters required for a high-performance multiplicity counter. Furthermore, non-^{3}He-based coincidence systems—equivalent to the HLNCC-II but based on LiZn/S scintillators and boron-coated proportional tubes—already have been demonstrated successfully (2014 ^{3}He Alternatives Workshop held at Joint Research Center in Ispra, Italy). Combining the DMM technique with these alternative neutron detection technologies suggests that MOX scrap measurements could be achieved without the need for any ^{3}He.

In this work, we have modeled the expected performance of the DMM technique for a variety of material properties. The models are supported by a limited measurement campaign validating the general approach to the measurement of the item multiplication without triples neutron counting. We have calculated the expected measurement precision as a function of plutonium mass, and examined potential biases for a number of material characteristics (i.e., U:Pu ratio, alpha, density, etc.) using the DMM technique. The expected measurement performance is of sufficiently high quality that a two-energy variant of the point model was implemented to take full advantage of the technique.

Analysis suggests that a simple neutron coincidence counter such as the HLNCC-II, coupled with the DMM methodology, will outperform the current state-of-the-art assay system, the Epi-Thermal Neutron Multiplicity Counter (ENMC), in terms of precision for equivalent assay times, over a broad range of material types. Measurement biases—even for extreme material types (e.g., α>10, [where α is the ratio of uncorrelated to correlated neutron emission] or U:Pu >10)—are expected to be on the order of 1% or less.

Safeguards relevance:

- The DMM is a viable alternative to neutron multiplicity analysis for MOX scrap assay.
- The DMM technique can be retrofitted to existing systems, extending their dynamic range.
- The DMM is compatible with already demonstrated alternative neutron detection systems. In principle, the DMM technique coupled with a PTI-based straw detector (as an example) could replace a traditional ^{3}He-based multiplicity counter.

Future Safeguards Technology Development Work:

- Although this work shows that DMM is extremely promising, it still must be field tested. A measurement campaign using MOX scrap materials is required to fully demonstrate the technique.
- We have introduced a U:Pu mass ratio correction to the DMM technique, which is presently a simple empirical model. Additional study is required to validate this correction. The validation can be performed via an MCNP exercise but would be improved by being tied into a direct field measurement.

1. INTRODUCTION

Neutron multiplicity counting is the most commonly used nondestructive assay technique for determining the plutonium mass within containers of scrap PuO_2 or mixed oxide (MOX). In multiplicity analysis, the $^{240}Pu_{eff}$ mass, leakage multiplication, and alpha ratio (the ratio of [α, n]-to-spontaneous fission neutron production) are the three primary unknown sample properties. They must be determined simultaneously. To solve for these three unknowns in a multiplicity assay, three measured values are needed: the singles, doubles, and triples neutron count rates. The need to determine the triples neutron count rate—combined with the high accidental coincidence background rates typical of these materials (due to the associated high [a, n] emission rates)—drives the need for very high neutron detection efficiency (>50%) and short neutron die-away times (<50 μs) in these applications. To achieve the necessary level of performance requires the use of large numbers (>100) of 2.54 cm diameter, high-pressure (>6 atm.) ^3He tubes committing 500 liters of ^3He (at standard temperature and pressure) or more for a single assay system. However, because of the limited supply and high cost of ^3He gas, these systems can no longer be considered cost-effective assay solutions.

The Direct Multiplication Measurement (DMM) [1] approach is an alternative neutron coincidence assay methodology that determines the sample's $^{240}Pu_{eff}$ mass, multiplication, and alpha ratio without the need for triples neutron counting. By adapting the Add-A-Source (AAS) matrix correction technique [2, 3] to the measurement of the multiplication, the remaining parameters, $^{240}Pu_{eff}$ mass and alpha ratio, can be determined from simple neutron coincidence counting analysis (i.e., known multiplicity, known alpha based on singles and doubles only). This measurement can be performed using more modest neutron counting systems, such as the High Level Neutron Coincidence Counter (HLNCC), which requires only a fraction of the ^3He needed compared to a multiplicity counting system [4], such as the Epi-Thermal Neutron Multiplicity Counter (ENMC).

Studies based on MCNPX simulations of the DMM using the HLNCC-II neutron coincidence counter have highlighted the limitations of the standard point model [4] used with traditional multiplicity analysis. Analysis of high-alpha samples using the standard point model can result in biases on the order of tens of percent. We have adopted a two-energy point model developed by Croft et al. [5] that effectively eliminates biases resulting from the standard point model.

2. ANALYSIS APPROACH

Multiplicity analysis is based on the point model [4], using three measured rates (single neutron rate [S] and double [D] and triple [T] neutron coincidences) to solve for three unknown properties of the sample:

$$S = m_{240} \cdot \Phi \cdot M \cdot \varepsilon \cdot \overline{\nu_{s1}} \cdot (1+\alpha) \ , \tag{1}$$

$$D = \frac{m_{240} \cdot \Phi \cdot M^2 \cdot \varepsilon^2 \cdot f_d}{2} \cdot \left\{ \overline{\nu_{s2}} + \left(\frac{M-1}{\overline{\nu_{i1}}-1} \right) \cdot \overline{\nu_{s1}} \cdot \overline{\nu_{i2}} \cdot (1+\alpha) \right\}, \text{and} \tag{2}$$

$$T = \frac{m_{240} \cdot \Phi \cdot M^3 \cdot \varepsilon^3 \cdot f_t}{6} \cdot \left\{ \overline{\nu_{s3}} + \left(\frac{M-1}{\overline{\nu_{i1}}-1} \right) \cdot \left[3 \cdot \overline{\nu_{s2}} \cdot \overline{\nu_{i2}} + \overline{\nu_{s1}} \cdot \overline{\nu_{i3}} \cdot (1+\alpha) \right] + 3 \left(\frac{M-1}{\overline{\nu_{i1}}-1} \right)^2 \overline{\nu_{s1}} \cdot (1+\alpha) \cdot \overline{\nu_{i2}}^{\,2} \right\}, \tag{3}$$

where

m_{240} = the $^{240}Pu_{eff}$ mass,
Φ = the spontaneous fission rate per gram ^{240}Pu,
M = the self-leakage multiplication,
E = the neutron detection efficiency,
f_d = the doubles gate fraction,
f_t = the triples gate fraction,
α = the ratio of uncorrelated to correlated neutron emission,
$\overline{\nu_{s1}}, \overline{\nu_{s2}}, \overline{\nu_{s3}}$ and $\overline{\nu_{i1}}, \overline{\nu_{i2}}, \overline{\nu_{i3}}$ = the spontaneous and induced fission prompt factorial moments, respectively.

The need to determine the triples rate to solve for the third unknown drives the need for a high-performance neutron multiplicity counting system (i.e., a system with high detection efficiency and a short die-away time) and its relatively huge ^{3}He inventory. With three measured rates, we can select three unknowns to solve for any of the following combinations of unknowns.

- Known efficiency: solve for m_{240}, α, M
- Known alpha: solve for m_{240}, ε, M
- Known multiplication: solve for m_{240}, α, ε
- Known mass: solve for M, α, ε

For MOX scrap assay, the neutron detection efficiency is assumed to be known and to be unaffected by the sample, so the known efficiency analysis is applied. However, if it is possible to determine the value of M, independently of the coincidence measurement, then the $^{240}Pu_{eff}$ mass and the α value can be determined from the singles and doubles rates alone; and the neutron detection performance requirements may be reduced. In this case, the $^{240}Pu_{eff}$ mass and α value are determined as follows:

$$m_{240} = \frac{1}{\varepsilon \nu_{s2} \Phi} \cdot \left[\frac{2D}{\varepsilon \cdot f_d \cdot M^2} + \frac{(M-1)}{M} \frac{\nu_{i2} S}{\nu_{i1} - 1} \right] ; \tag{4}$$

$$\alpha = \frac{S}{\varepsilon \nu_{s1} M \Phi m_{240}} - 1 . \tag{5}$$

Note that Krick et al. have previously noted that the standard point model approach does not adequately represent the more challenging plutonium material forms such as metals [6]. We found that the standard point model is also not suitable for the scrap MOX materials of interest here. A use of a two-energy point model, developed by Croft et al. [5] will be discussed in Section 7.2 since this method eliminates biases resulting from the standard point model.

3. MEASUREMENT APPROACH

Measurement systems such as the Active Well Coincidence Counter (AWCC) [7] and ^{252}Cf shuffler [8] are used to determine the ^{235}U content of a sample through active neutron interrogation. For the active well, neutrons emitted from fixed Am(Li) neutron sources induce fission within the sample, and the resulting fission neutrons are detected via neutron coincidence counting. The shuffler technique relies on the detection of delayed neutrons from fission events induced by the brief introduction of a ^{252}Cf source into the assay cavity with the sample. The neutron count rates from both of these techniques are used to

determine the ^{235}U mass within the sample. However, in both cases, the observed count rates are more closely related to the sample's self-multiplication than to its mass. In these systems, we measure the increase in induced fission rate due to an external neutron source (i.e., the multiplication), from which a fissile mass is inferred via an empirical calibration. (It is because these systems do not directly measure the fissile mass that they must be calibrated with representative standards; and sample deviations from the standard's composition, density, and geometry can result in significant biases in the reported mass value.) The DMM applies a similar active neutron interrogation to directly measure the sample multiplication.

It would seem that either the shuffler or active well techniques could be used to measure the sample's self-multiplication directly for use in the point model analysis. However, an evaluation of these two obvious candidate technologies, the ^{252}Cf shuffler and the traditional AWCC, shows that neither is particularly practical for this application. The Am(Li) sources used with the traditional AWCC have neutron emission rates that are too low, and the sources are physically too large to be easily installed in and removed from the assay cavity during the course of an assay. Additionally, Am(Li) sources are subject to supply limitations and are difficult to obtain. The typical ^{252}Cf shuffler is a very large, costly assay system and so is not a convenient measurement platform for routine use.

In this work, the measurement is performed in a manner similar to the AAS [3] matrix correction for neutron coincidence assay of waste drums. A small (1×10^5 to 1×10^6 n/s) ^{252}Cf source is introduced into the neutron detector assay cavity as near to the sample as is practical, as shown in Fig. 1. The AAS method for correcting the Pu doubles rate is accomplished by measuring the effect the matrix has on the count rate of an external ^{252}Cf source. Since the matrix effect of scrap MOX powders is generally negligible, the impact of the sample on the net ^{252}Cf coincidence rate should be a simple function of the sample's self-multiplication.

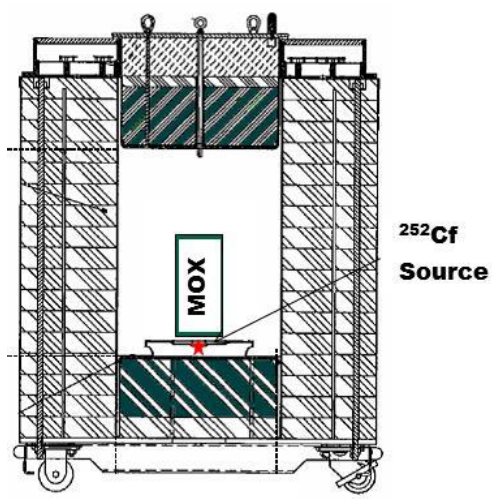

Fig. 1. Illustration of the source location for the ^{252}Cf source used for the Direct Measurement of Multiplication measurements.

The source may be positioned manually or by using an automated source transfer mechanism such as that found in the shuffler or the waste drum assay system with an AAS correction module [3]. The measurement is repeated with and without the ^{252}Cf in the assay cavity. The sample container is first assayed alone within the assay cavity; the ^{252}Cf source is then added and the measurement is repeated. The relative net coincidence doubles rate ratio, *R*, is determined as

$$R = \frac{D_{Pu+Cf} - D_{Pu}}{D_{Cf}}, \tag{6}$$

where

D_{Pu+Cf} = the doubles rate with sample in place and ^{252}Cf source in the irradiated position,
D_{Pu} = the doubles rate from the passive neutron coincidence measurement of the sample,
D_{Cf} = the doubles rate from the passive neutron coincidence measurement of the ^{252}Cf source

NOTE: All neutron count rates are corrected for dead-time.

The self-multiplication is expected to be a simple function of the ^{252}Cf doubles rate ratio, $M = M(R)$.

MCNPX [9] modeling was employed to examine the relationship of the sample's multiplication to the change in the ^{252}Cf coincidence rate. As an example, a model of a typical multiplicity counting system (the plutonium scrap multiplicity counter [PSMC]) was created; and the measurement response as a function of fill height (0 to 24 cm in 2 cm increments) was calculated for two different bulk densities of MOX, both with and without the ^{252}Cf source present. The multiplication for the sample was determined for each combination of mass and density, and then the predicted doubles (Reals) rates for a ^{252}Cf source located directly beneath the container were determined. The MOX materials were defined as having bulk densities of either 2.5 g/cc or 3.5 g/cc, a U:Pu ratio of 2:1, and $^{240}P_{eff}$ of 0.38 g/g. The model suggests a surprisingly linear relationship between the multiplication and the relative doubles rate from the ^{252}Cf (Fig. 2). Based on this result, the sample multiplication is given as

$$\mathrm{M} = \mathrm{a} \cdot \mathrm{R} + \mathrm{b} = \mathrm{a} \cdot \left[\frac{D_{Pu+Cf} - \mathrm{D_{Pu}}}{\mathrm{D_{Cf}}}\right] + \mathrm{b}\,, \tag{7}$$

where *a* and *b* are constants specific to a given assay system and container size.

This relationship is expected to hold, provided the container size and location within the cavity are consistent and the sample matrix is relatively homogenous. Unexpected deviations from the calibration, such as voids, are expected to adversely impact the performance, perhaps more strongly than would be expected for a traditional multiplicity or AWCC measurement.

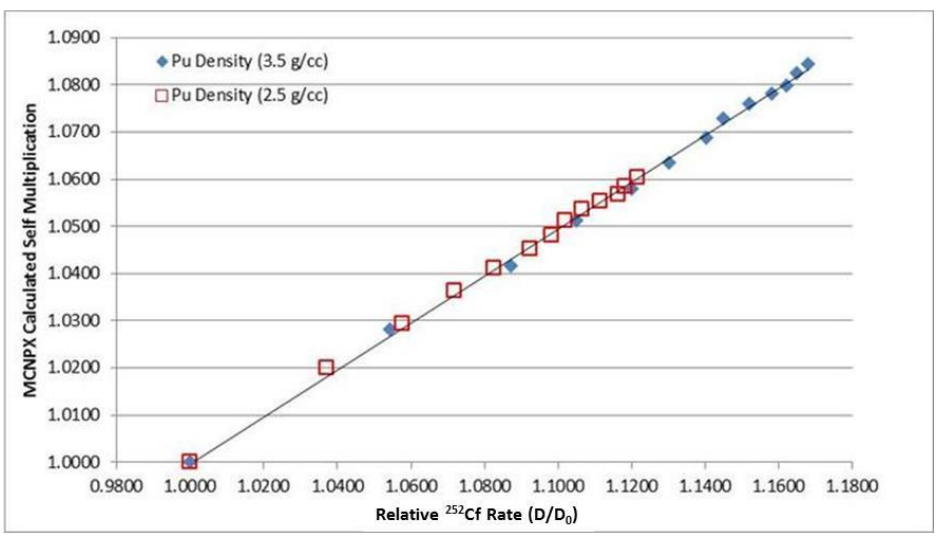

Fig. 2. Plot showing the expected relationship between the sample self-multiplication as a function of the relative ^{252}Cf rates for the Direct Measurement of Multiplication assay with a PSMC. These data represent the multiplication and Add-A-Source rates for two different densities of PuO_2 for a series of increasing masses of PuO_2 within a 9 cm diameter container. R_0 represents the ^{252}Cf Reals rate with no sample present in the assay cavity.

4. EARLY ATTEMPT AT THE DMM TECHNIQUE

A previous attempt at the DMM technique was made several years ago [10], but it was executed in a very informal way taking advantage of an opportunity to assay a number of high-burnup MOX scrap containers. Because this was an impromptu experiment, we made do with materials on hand.

Those measurements were performed using a standard PSMC, which provides 54% detection efficiency with 50 µs die-away time. Data acquisition was performed using a JSR-14 multiplicity shift register operated using the Los Alamos National Laboratory INCC software. The PSMC operated as a multiplicity counter was the primary assay method for scrap MOX at this facility. For these initial measurements, we had only a 30,000 n/s ^{252}Cf source, which limited the measurement precision achievable for the DMM testing.

Several MOX samples were made available to us for assay with the PSMC. These samples ranged in mass from approximately 100 to 1200 g of Pu with a nominal Pu-240 $_{\text{effective}}$ mass per gram of 0.27. The alpha values (indicator of the alpha-n emission rate) of these samples ranged from 5 to greater than 20. The neutron yields from some of these samples exceeded 1×10^6 n/s. (Note: These samples were not standards; the mass values used for examination of the DMM approach were obtained from the multiplicity assay.)

Each MOX sample was measured both with and without the ^{252}Cf source. (We were also investigating the possibility of using a ^{252}Cf-based AWCC measurement for MOX, so the ^{252}Cf source was alternatively placed at the top and the bottom of the chamber.) The typical measurement time available was approximately 2 to 2.5 hours per container, and the measurement time was divided approximately so that about 1 hour was used for active measurements and 1 hour for the passive (without ^{252}Cf) measurements. Multiple measurements were made of each sample as time and sample availability allowed. The measurement arrangement is shown in Fig. 3.

For each MOX sample assay, the measurement sequence was as follows:

- MOX sample assayed alone
- ^{252}Cf source measured at the bottom of the chamber with the MOX sample in place
- ^{252}Cf source measured at the top of the chamber with the MOX sample in place

In addition, reference measurements were made with the ^{252}Cf source positioned at the bottom then the top of the assay cavity at the beginning and the end of the measurement campaign (which could span from a few hours to up to a week in duration).

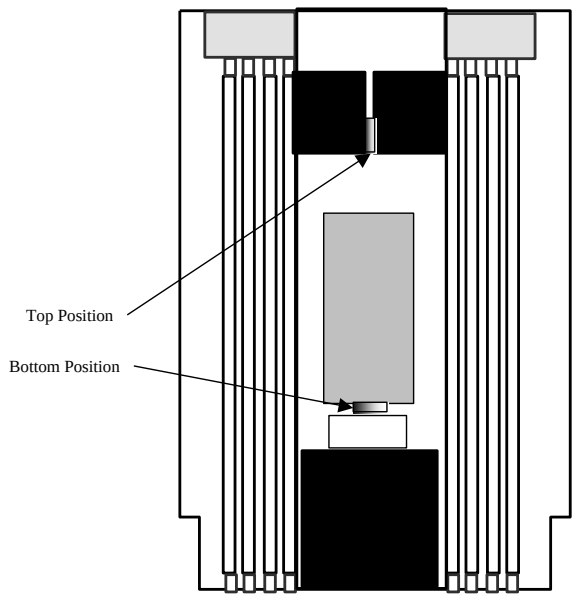

Fig. 3. Sketch showing the measurement geometry of the early trials using a plutonium scrap multiplicity counter with mixed oxide samples.

Because we had the benefit of performing this study with a high-performance neutron multiplicity counter, we were able to determine each sample's self-multiplication from the measured rates without the need for calibrated standards or an MCNP-based study. We consider the change in the sample multiplication in terms of the samples' impact ^{252}Cf source. Figure 4 presents a plot of the measured multiplication (from the multiplicity analysis) as a function of the observed change in relative ^{252}Cf count rate ratios (also called the ^{252}Cf perturbation factor). The plot is seen to be linear, but the deviation about the line is large compared with the measurement precision. (Note that the error bars representing the measurement precision are shown in the plot but are so small that they are indistinguishable from the plotted points themselves).

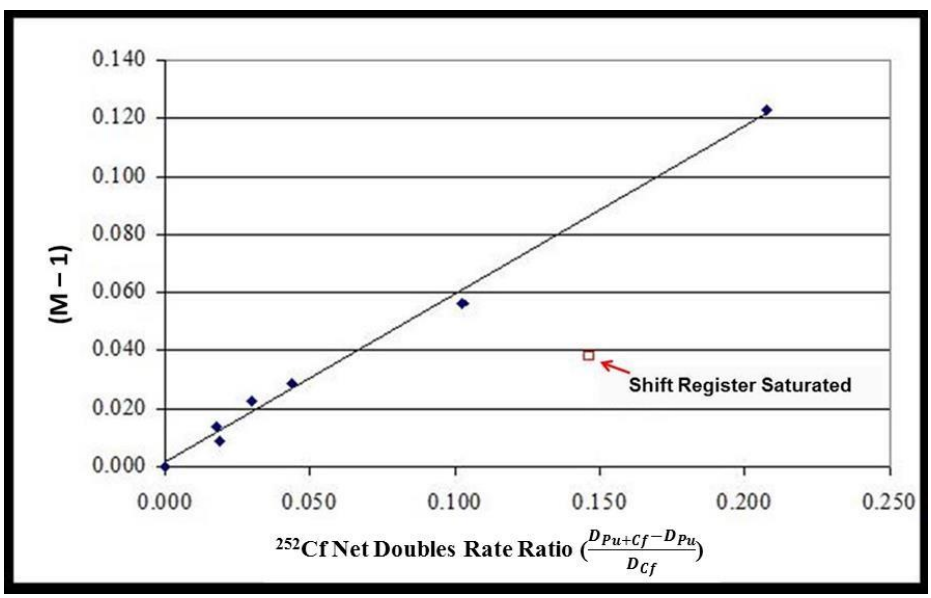

Fig. 4. Plot of the scaled measured multiplication, M, (determined from multiplicity analysis) as a function of ^{252}Cf source perturbation. For the "hottest" mixed oxide sample, count rates in excess of 1 MHz resulted in saturation of the now-outdated JSR-14 shift register.

The measurements were evaluated both using the standard multiplicity analysis using singles, doubles, and triples rates, and using the DMM method considering only the singles and doubles. The resulting measured mass values (normalized to the declared mass) are plotted against the declared values in Fig. 5, which shows that the DMM and PSMC multiplicity analyses performed similarly. However, for these samples, the declared mass values were determined by a prior multiplicity assay using the PSMC; therefore, one might expect the PSMC results to compare well to the declared values.

The early measurements were hampered by the following conditions:

- The ^{252}Cf source was too small relative to the MOX sample neutron yields, adversely impacting measurement precision.
- The ^{252}Cf source positioning was not reproducible because source positioning was done manually.
- The MOX sample positioning was not reproducible because no centering device was available.
- The multiplicity mass assay of the high-alpha MOX samples is known to produce biases, complicating the evaluation of the DMM results
- Assay time was limited.

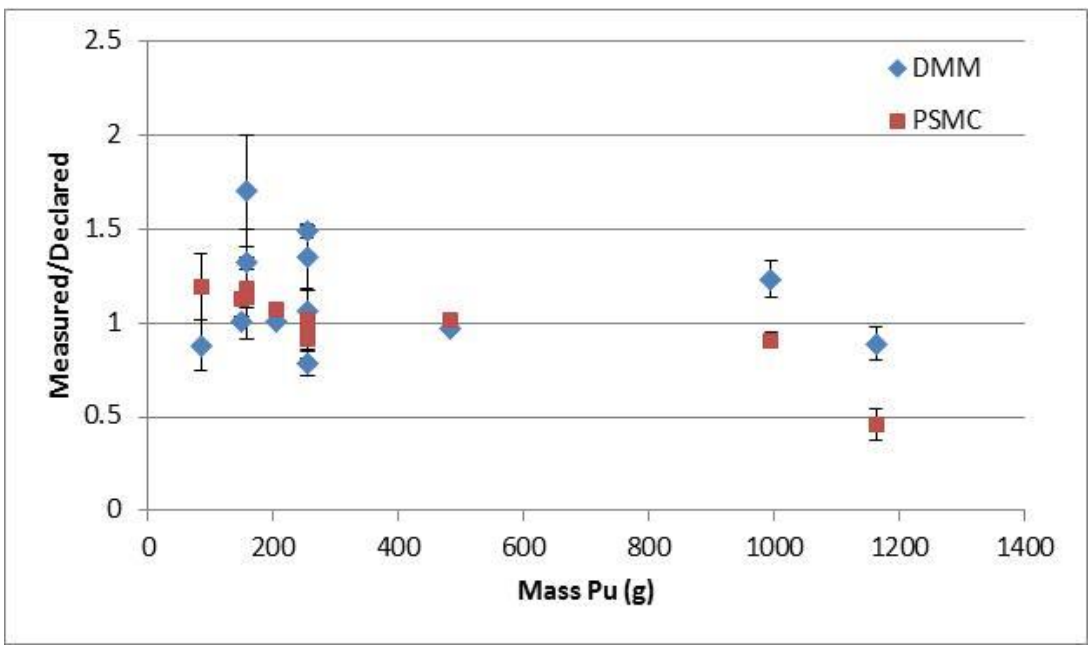

Fig. 5. Comparison of the Direct Measurement of Multiplication and traditional multiplicity analysis results using the plutonium scrap multiplicity counter.

These early measurements of high-burnup scrap MOX containers suffered from a poor signal-to-noise ratio due to inadequate ^{252}Cf source strength and uncertainties in repositioning the ^{252}Cf source. The results, although promising, were inconclusive. However, it was encouraging to note that these experiments, with simple coincidence measurements using only the singles and doubles rates, were able to achieve results comparable to those obtained with the full multiplicity analyses.

5. EXPERIMENTAL CONFIRMATION (CURRENT STUDIES)

5.1 ^{252}Cf Shuffler Measurements

In this work, our goal was to evaluate the feasibility of this simple, alternative measurement technique in an optimal measurement configuration with minimal interference issues. The intent was to use Oak Ridge National Laboratory's ^{252}Cf shuffler to provide a mechanically reproducible placement of a high-yield neutron interrogation source to examine the induced fission response from a series of well-characterized ^{235}U sources for comparison with MCNPX simulated values. Although the shuffler is physically much larger than the HLNCC-II, the two instruments have similar neutron coincidence performance metrics (efficiency 17.6%, die-away 50 μs). Uranium-235 was to be studied, rather than plutonium, to provide a low passive background that would facilitate comparison of predicted and modeled performance.

However, the neutron source rate for the shuffler was very high (~2×10^7 n/s), requiring the installation of a derandomizer circuit to minimize the dead time. In addition, the source transfer mechanism was found to have significant positioning uncertainty. The source positioning errors pushed uncertainties to as high as 0.6% in the singles rates (Fig. 6) and 1.2% in the doubles rates. These non-statistical variations in count rate were too large to tolerate for the multiplication measurement. These results highlight the importance of reproducible source positioning. While these issues were addressed, an alternative assay system, the Large Volume AWCC (LAWCC), was pressed into service for the measurements.

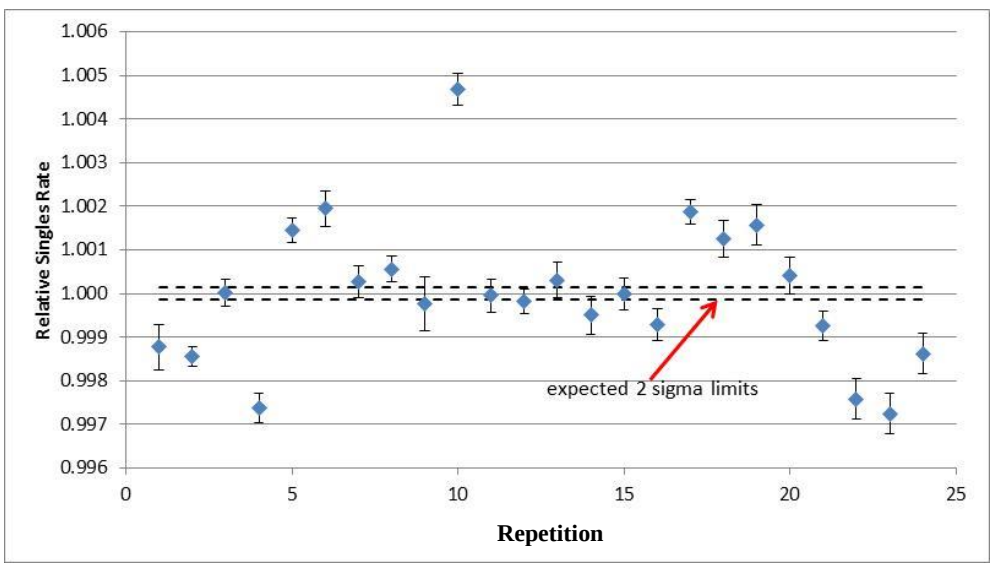

Fig. 6. Plot showing the scatter in the observed count rate from the shuffler due to jitter in the source position.

5.2 Large Volume Active Well Coincidence Counter

The LAWCC is similar in design to the standard AWCC but has been scaled to a larger cavity diameter [7]. It incorporates 48 ^{3}He proportional tubes arranged in two concentric rings around an 28cm diameter, 38 cm tall assay cavity. Each tube measures 1 in. in diameter and is filled to a partial pressure of 4 atm. For these measurements, the high-density polyethylene end-plugs were replaced with graphite end-plugs

to more closely simulate a typical, passive-neutron coincidence counter. The neutron detection efficiency and die-away time for the LAWCC are 34% and 50 µs, respectively. The general layout of the LAWCC can be seen in Fig. 7.

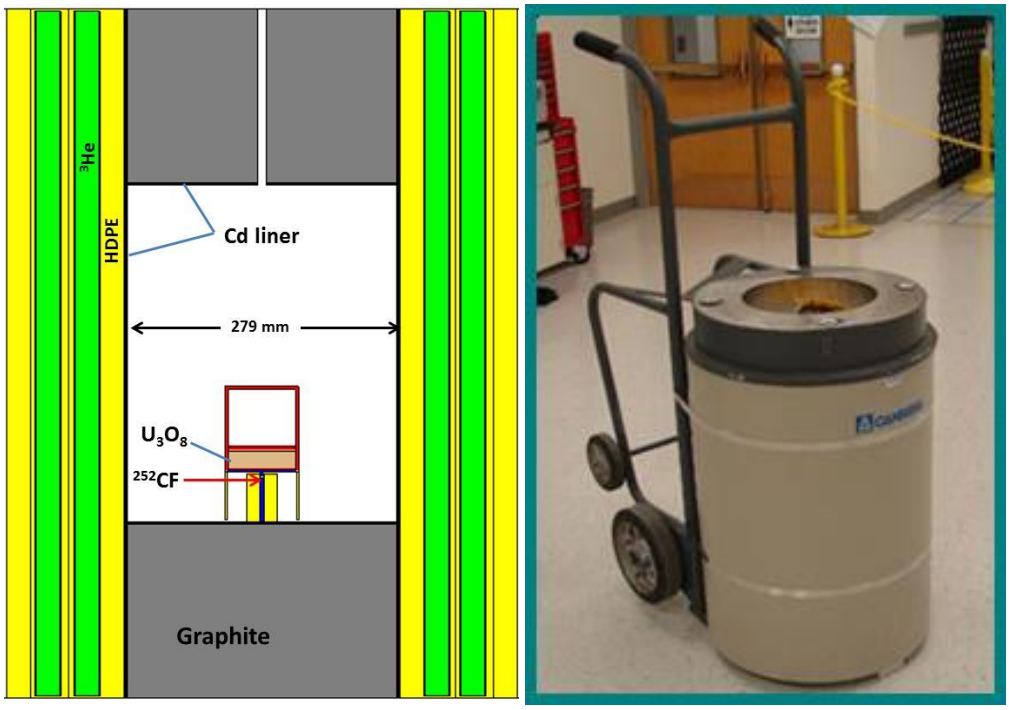

Fig. 7. Screen shot of the MCNPX input file for the Large Volume Active Well Coincidence Counter (LAWCC) configured for the Direct Measurement of Multiplication test run (left) and a photograph of the LAWCC (right).

To examine the linearity and measurement precision possible for the DMM measurement, a series of containers of enriched uranium oxide were assayed in the LAWCC to determine the californium source ratio, *R*. The measurement geometry was modeled using MCNPX to provide a leakage multiplication value for each container. The highly enriched uranium (HEU) items (New Brunswick Laboratory Certified Reference Material 146 [11]) each contained approximately 200 g of uranium at various enrichments in the form of U_3O_8 powder (Table 1). The ^{252}Cf source used for these measurements was approximately 60 ng with a neutron yield of 1.4×10^5 n/s. The observed singles and doubles rates are shown in Fig. 8.

Table 1. Uranium standards from National Bureau of Standards and New Brunswick Laboratory (NBL) measured in the LAWCC

Standard description	^{235}U enrichment	Total mass ^{235}U (g)	Total mass U (g)
NBL-0001	20.11	39.10	194.43
NBL-0002	52.49	101.72	193.79
NBL-0003	93.17	181.15	194.43

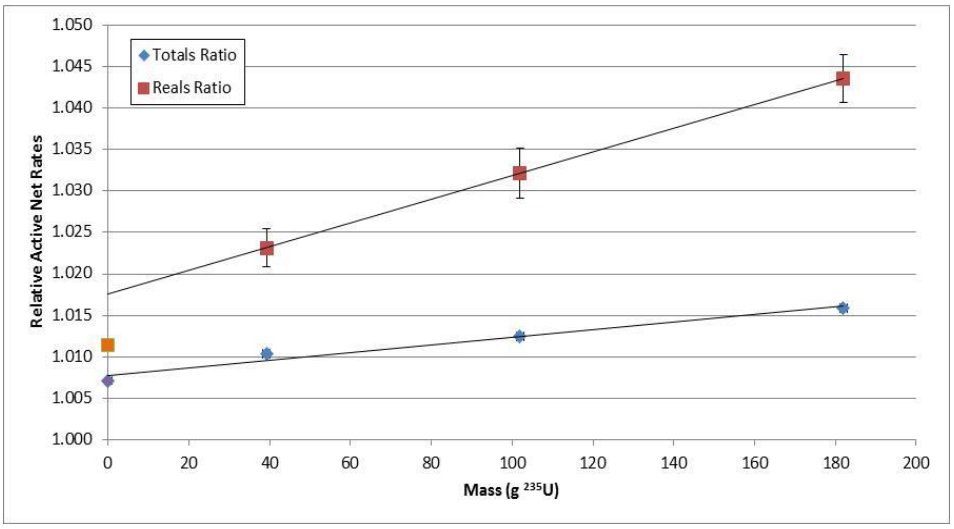

Fig. 8. Measured Totals neutron count rate and Reals rate ratios for the New Brunswick Laboratory U_3O_8 cans measured in the modified Active Well Coincidence Counter as a function of ^{235}U mass. The *m* = 0 point is the New Brunswick Laboratory blank container.

Fig. 9 shows the observed count rate ratios as a function of multiplication (calculated from MCNPX) results in linear fits for the U_3O_8 containers, lending credibility to the MCNPX calculations for the MOX containers. Repeat measurements were performed on these HEU containers to estimate the impact of sample positioning on the assay results. Additional measurements were performed on the containers deliberately loaded into the assay cavity at an offset from the centerline by 1.27 cm. Replicate measurements with hand loading of the sample resulted in a standard deviation in the relative ^{252}Cf rates of <0.2%. The offset measurements caused a 0.3% average bias in the assay result.

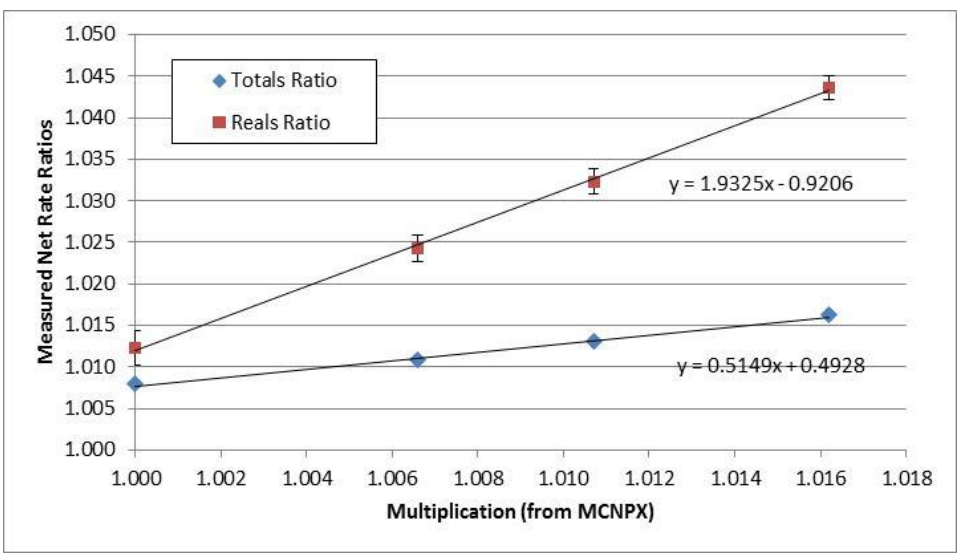

Fig. 9. Measured total and Reals rate ratios for the New Brunswick Laboratory U_3O_8 cans (listed in Table 1) measured in the modified Active Well Coincidence Counter as a function of the MCNPX-calculated self-multiplication for each standard.

6. MEASUREMENT PRECISION FOR MOX ASSAY USING THE DMM TECHNIQUE

In this section, the measurement precision for MOX samples is estimated for a 1 hour multiplicity assay measurement in the ENMC and for a 1 hour neutron coincidence measurement in an HLNCC-II, divided evenly between the passive sample assay and the active sample assay with ^{252}Cf , respectively. We first examine the precision of the multiplication measurement using the DMM/HLNCC-II technique compared with the measurement using a full multiplicity analysis in an ENMC (MULT/ENMC). The plots in Fig. 10 provide the expected precision in the determination of the sample self-multiplication as a function of a MOX mass packaged within a typical 15 cm diameter container using a 0.5 µg ^{252}Cf interrogating source. Fig. 10 shows that, for the material range of most interest [1 < g Pu < 1000, and $\alpha > 1$], the DMM technique should provide a more precise value for the reported Pu mass.

The estimated measurement precision of an HLNCC-II with the DMM technique for $^{240}Pu_{eff}$ mass is calculated based on the directly measured value for multiplication and is compared with the calculated measurement precision for multiplicity analysis using the ENMC. For this example, we again assume a 0.5 µg (1×10^6 n/s) ^{252}Cf source and a total assay time of 3600 s for the two measurements. For the DMM approach, the assay is divided into 1800 s passive and active assays. Fig. 11 provides a summary of the estimated measurement precision achievable with the DMM technique. The measurement performance for the HLNC, coupled with the DMM technique, is expected to equal or exceed the observed performance of the standard ENMC for the expected range of quantities in MOX powders and MOX scrap materials other than product grade materials.

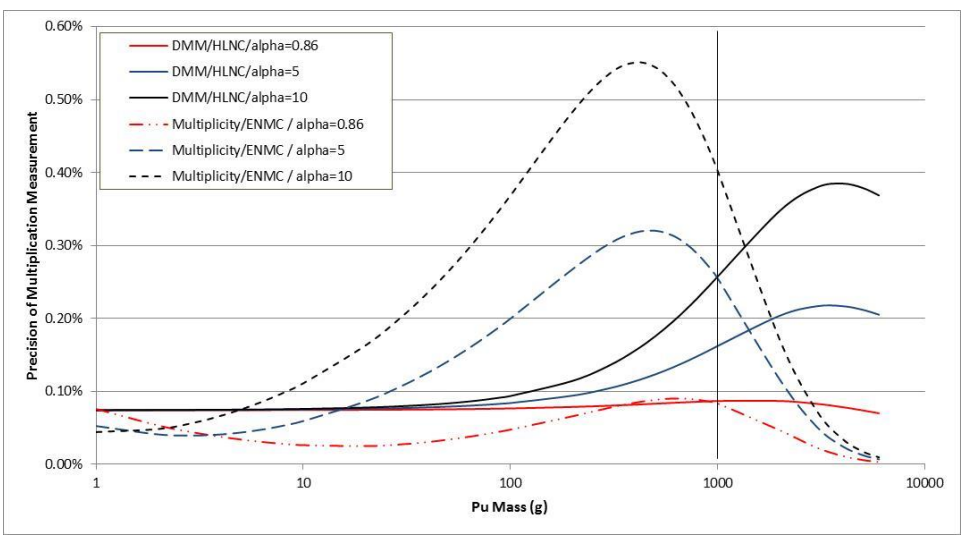

Fig. 10. Expected measurement precision in the reported multiplication for multiplicity analysis using the ENMC (63% neutron detection efficiency) and for the Add-A-Source style multiplication measurement using a standard HLNCC-II (18% efficiency) and a 1×10^6 n/s ^{252}Cf interrogation source.

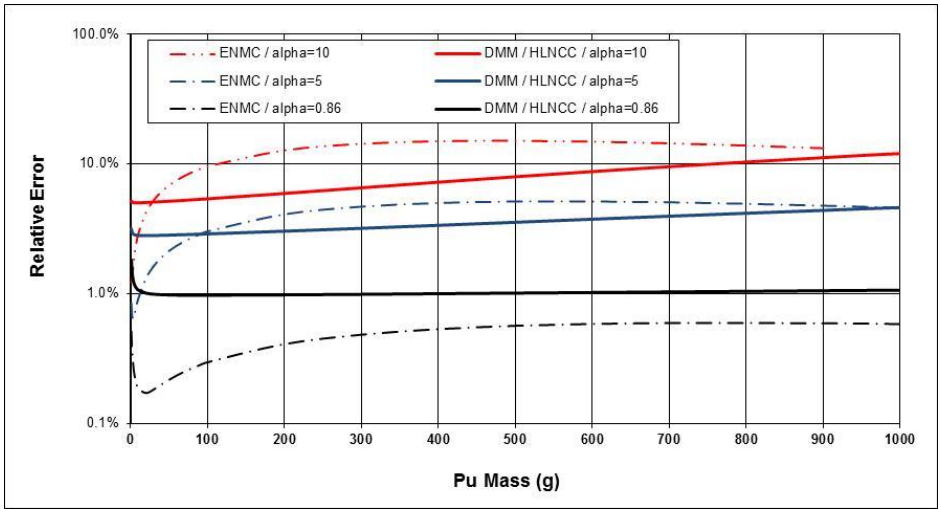

Fig. 11. Expected measurement precision in the reported plutonium mass for multiplicity analysis using the ENMC (63% neutron detection efficiency) and for the Add-A-Source style multiplication measurement using a standard HLNC (18% efficiency).

7. ACCURACY OF THE DMM TECHNIQUE FOR MOX ASSAY

Measurement performance must also consider potential biases as well as precision. To evaluate the potential accuracy of the DMM technique, we chose to use Monte Carlo modeling to examine the performance for MOX materials assayed using an HLNCC-II. The measurement response as a function of density, mass loading, isotopic abundance and U:Pu ratio was examined using MCNPX. (Note: these factors also impact the accuracy of the traditional multiplicity assay.)

7.1 BASELINE MCNPX MODEL

The HLNCC-II MCNPX model (Figure 12) was derived from the Canberra Industries HLNCC specification sheet for the JCC-31 HLNCC-II [12]. Basic performance parameters for the model compared well with the measured values for a commercial off-the-shelf HLNCC. The simulated detection efficiency in this system for a ^{252}Cf point source was 17.9%, compared with a 17.8% measured efficiency, and the die-away time was 41 μs simulated vs. 42 μs measured. Because of the time constraints of this study, models were produced only for a single container size (13 cm inner diameter by 30 cm height).

Initial simulations examined the basic mass dependence of the DMM approach for a stoichiometrically pure (oxygen to metal ratio = 2) MOX composition, with α= 1, 3 , and 10 and $^{240}Pu_{eff}$ mass of 0.389 per gram of plutonium.

Simulation of the Measurement Response

The measurement response was determined by the summation of three separate iterations of the MCNPX model using different neutron source terms.

- Fission signal—represents the neutron signal resulting from interactions initiated by the spontaneous fission of the plutonium within the MOX container. In this case, a uniform source of neutrons is distributed throughout the sample volume with energy distribution representative of a ^{240}Pu fission spectrum. The resulting simulated signal includes both the spontaneous fission signal and the fission signal induced by the passive neutron source term.

- (α, n) Signal—represents the neutron signal from interactions initiated by neutrons that result from the interaction of alpha particles on the low-Z constituents of the sample matrix. The neutron energy distribution was estimated using the Sources4C program [13]. The resulting simulated signal includes both the neutron rates from the (α, n) source term and the fission signal induced by the (α, n) source term.
    - The (α, n) contribution was multiplied by an arbitrary value to provide various levels of alpha for a given isotopic composition.
- Active ^{252}Cf interrogation—a point source of ^{252}Cf spontaneous fission neutrons located just below the MOX sample container. The resulting simulated signal provides the coincidence rates for the multiplication measurement.

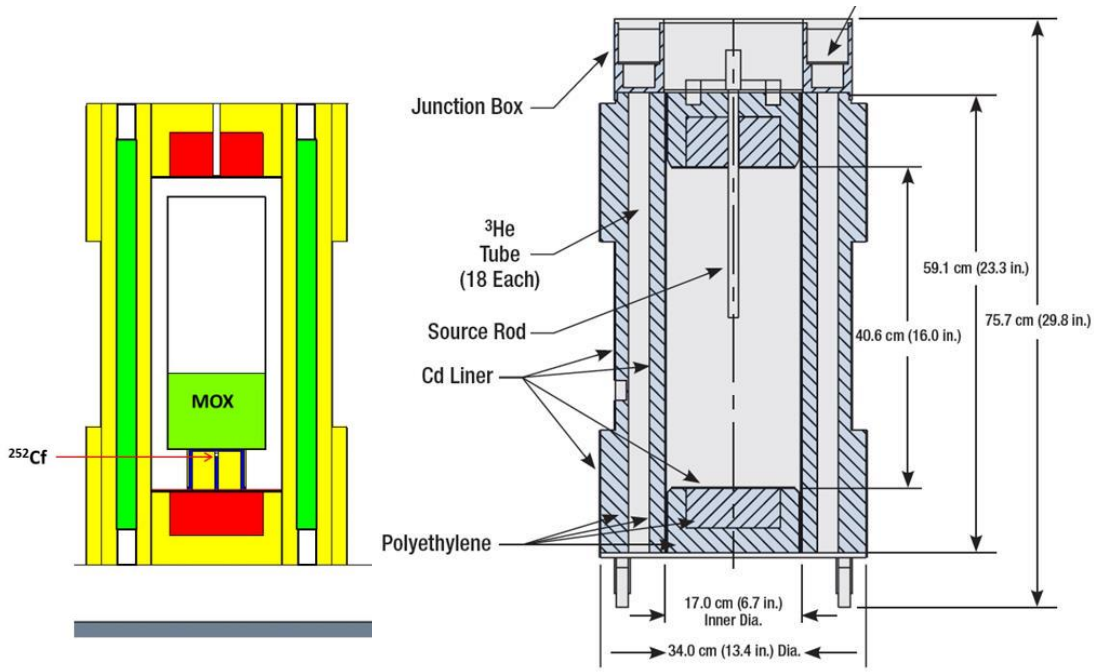

Fig. 12. Screen shot of the MCNPX input file used for the bias and precision studies (left) and a sectioned drawing of the HLNCC (right). Note: The electronics junction box (at the top of the section drawing) was not included in the model.

7.2 TWO ENERGY POINT MODEL

The bias depicted in Fig. 13 is not unique to the DMM approach; but it is a limitation of the point model, which assumes that induced fission events can be described by a single isotopic fission neutron energy spectrum (e.g., ^{240}Pu) and a single distribution of multiplicity moments (e.g., characteristic of ^{239}Pu). However, as the value of alpha increases, the neutron energy distribution changes, and the model must incorporate the changes in reaction and detection due to the (α, n) energy distribution—which the standard point model does not do.

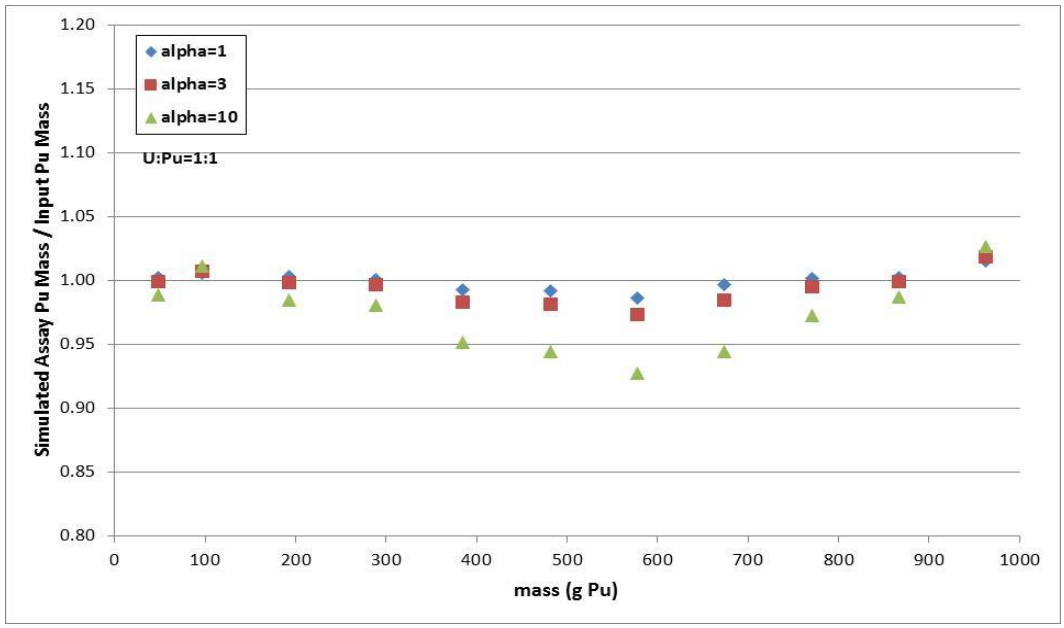

Fig. 13. Expected measurement performance of the Direct Measurement of Multiplication technique integrated with the standard point model (α = 1, 3, and 10 and U:Pu=1:1). The plots show the ratio of the mass calculated from the modeled count rates to the plutonium mass input to the MCNPX simulation. The deviation between the resulting simulated measurement and the input value results is caused by a limitation of the point model (this limitation is described in Section 7.2).

We have used the Two Energy Point Model developed by Croft [5] to address this bias. The Two Energy Point Model is as follows:

$$S = \varepsilon \Phi m_{240} \nu_{S1} M \left[1 + \alpha \left(\frac{a_p (M-1)\nu_{\alpha 1} + a_\varepsilon \left[(\nu_{I1} - a_p) + (a_p - 1)/M \right]}{M\nu_{I1} - 1} \right) \right] \tag{8}$$

$$D = \varepsilon^2 f \Phi m_{240} M^2 \left[\frac{\nu_{S2}}{2} + \left(1 + \alpha a_p \left(\frac{(M-1)\nu_{\alpha 1} + (\nu_{I1} - 1)\nu_{\alpha 2}/\nu_{I2}}{M\nu_{I1} - 1} \right) \right) \nu_{S1} \left(\frac{M-1}{\nu_{I1} - 1} \right) \frac{\nu_{I2}}{2} \right] \tag{9}$$

where

a_ε	=	the ratio of the detection efficiency (count per neutron from neutrons created in [α, n] reactions) to the detection efficiency from neutrons created in fission processes that emerge from the item. This value can be determined by MCNP or measurements by differentiating between singles and coincidence neutrons.
a_p	=	the ratio of the probability that a neutrons created in an (α,n) reaction will induce fission in the item to the corresponding probability for neutrons created in fission. This value can be determined by MCNP or measurements.
$\nu_{\alpha 1}$ and $\nu_{\alpha 2}$	=	the first and second factorial moments of the induced-fission prompt-neutron multiplicity distribution resulting from (α,n) neutron interactions

Assuming one knows or has measured the multiplication value, these equations may be solved for the $^{240}Pu_{eff}$ mass and alpha.

$$m_{240} = \left(D - \frac{c_2 c_7}{c_1} S \right) / (c_6 - c_2 c_7)/\Phi \,. \tag{10}$$

$$\alpha = \left(\frac{S}{c_1 \cdot m_{240} \cdot \Phi} - 1\right) \cdot c_2 \ . \tag{11}$$

where

$$c_1 = \varepsilon \nu_{S1} M \ ,$$
$$c_2 = \frac{M\nu_{I1} - 1}{a_p(M-1)\nu_{\alpha 1} + a_\varepsilon[(\nu_{I1}-1) + (a_p - 1)/M]} \ ,$$
$$c_3 = \varepsilon^2 f M^2$$
$$c_4 = a_p \left(\frac{(M-1)\nu_{\alpha 1} + (\nu_{I1}-1)\nu_{\alpha 2}/\nu_{I2}}{M\nu_{I1} - 1}\right) ,$$
$$c_5 = \nu_{S1} \left(\frac{M-1}{\nu_{I1}-1}\right) \frac{\nu_{I2}}{2} \ ,$$
$$c_6 = \frac{\nu_{S2}}{2} c_3 + c_3 c_5 = \varepsilon^2 f M^2 \left[\frac{\nu_{S2}}{2} + \nu_{S1} \left(\frac{M-1}{\nu_{I1}-1}\right) \frac{\nu_{I2}}{2}\right] ,$$
$$c_7 = c_3 c_4 c_5 = \varepsilon^2 f M^2 a_p \left(\frac{(M-1)\nu_{\alpha 1} + (\nu_{I1}-1)\nu_{\alpha 2}/\nu_{I2}}{M\nu_{I1} - 1}\right) \nu_{S1} \left(\frac{M-1}{\nu_{I1}-1}\right) \frac{\nu_{I2}}{2}$$

Although this representation seems somewhat complex, the only additional information required to solve these equations—relative to the simple point model—is a general knowledge of the type of impurity driving the high (α ,n) reaction rate (e.g., oxygen, fluorine, or beryllium). Reanalyzing the data set used to create Fig. 13 with the Two Energy Point Model results in lower bias, as shown in Fig. 14. Based on the (a, n) distribution in the work of Jacobs and Liskien [14], there is only a small difference between the average energies of the (a, n) on oxygen and the spontaneous fission neutron distribution from ^{240}Pu. To properly illustrate the need for correction, Fig. 15 shows the performance using the somewhat higher energy distribution based on the Sources 4C program (average neutron energy from the (α, n) reaction is 2.36 MeV).

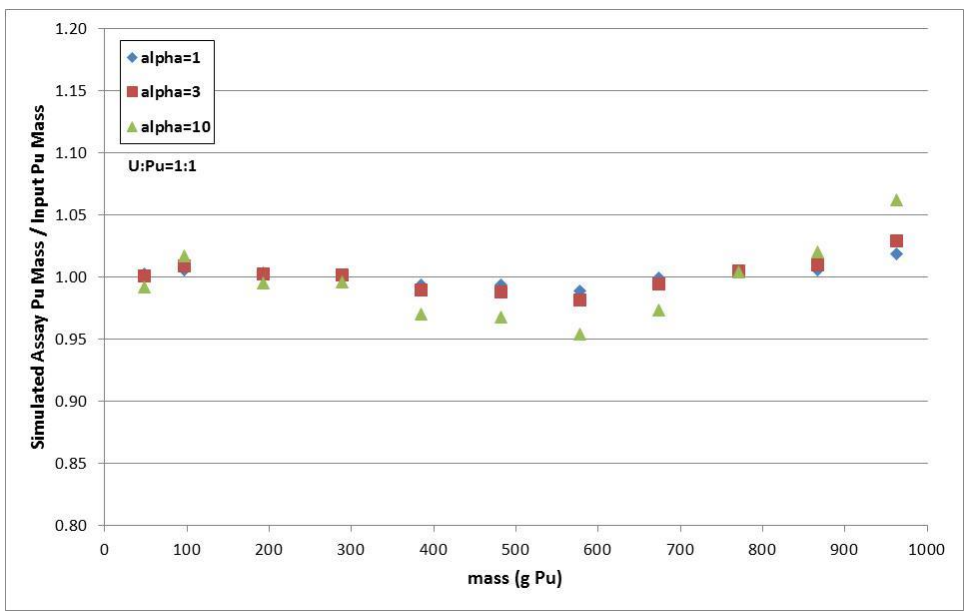

Fig. 14. Expected measurement performance of the Direct Measurement of Multiplication technique integrated with the two energy point model (α = 1, 3 and 10; U:Pu=1:1). The plots show the ratio of the mass calculated from the modeled count rates to the plutonium mass input into the MCNPX simulation. The deviation between the resulting simulated mass measurement and the input value results is caused by a limitation of the point model.

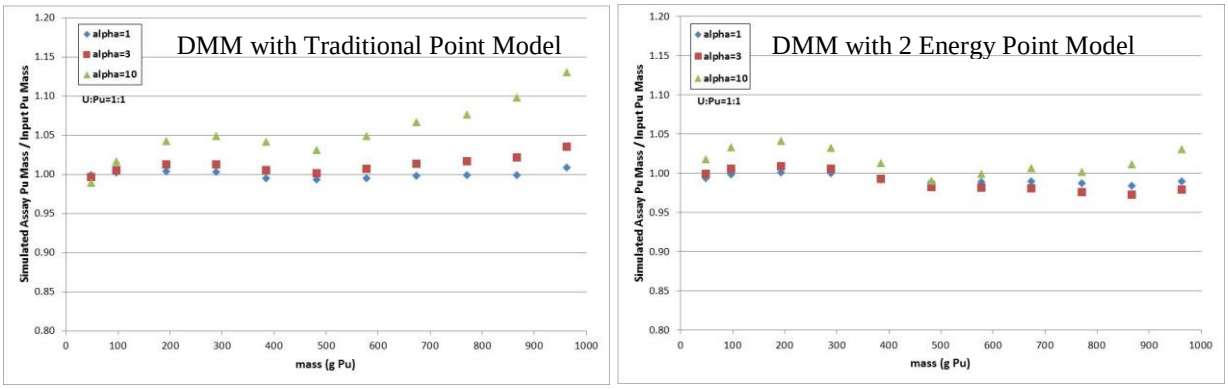

Fig. 15. Expected measurement performance of the Direct Measurement of Multiplication technique integrated with the two energy point model (α = 1, 3 and 10; U:Pu=1:1) using the Source 4C energy (α, n) distribution. The plots show the ratio of the mass calculated from the modeled count rates to the plutonium mass input into the MCNPX simulation. The deviation between the resulting simulated mass measurement and the input value results is caused by a limitation of the point model.

7.2.1 Determination of the Parameters a_ε and a_p

As mentioned above, the parameter a_ε in Eq. 8 can be estimated using the MCNPX models for the HLNCC-II. Figure 15, provides the expected neutron detection efficiency as a function of energy for a point source positioned at the center of the HLNCC-II assay cavity. The solid line in Fig. 16 represents the neutron detection efficiency for a series of mono-energetic point sources. The efficiency for three isotopic point sources with distributed neutron energy (ie. ^{240}Pu spontaneous fission neutrons, ^{252}Cf spontaneous fission neutrons and the O(α, n)Ne reaction neutron distributions). From these results based on neutron energy distributions determined from the work of Jacobs and Liskien [14], we estimate the value of the parameter a_ε to be 0.942 and to be 0.93.

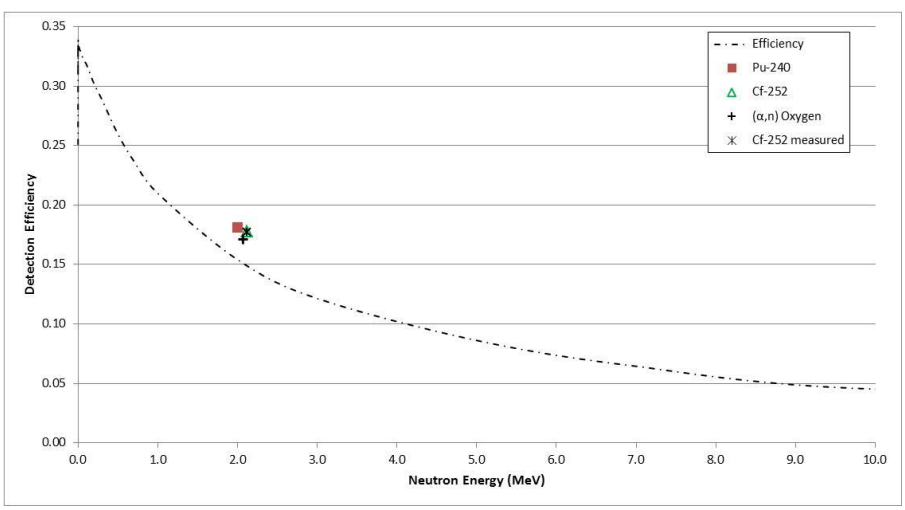

Fig. 16. Neutron detection efficiency as a function of energy for the HLNCC-II for a mono-energetic point source in the center of the HLNCC-II assay cavity. Overlain on the graph are the detection efficiencies for point sources of ^{240}Pu, ^{252}Cf and an oxygen (α, n) source. The measured efficiency for a ^{252}Cf point source [15] is plotted for comparison.

7.3 IMPACT OF THE U:PU RATIO ON THE MULTIPLICATION MEASUREMENT

A series of simulations for increasing plutonium masses were performed for three different values of U:Pu (the uranium was taken as natural) to examine the impact of multiplication on the DMM measurement and on the subsequent mass determination (Fig. 17). The simulations revealed that the DMM response (as a function of self-multiplication) has a significant dependence on the U:Pu ratio such that the simple response function discussed in Section 3 does not yield an accurate multiplication for use with the point model (Fig. 18). This limitation is due primarily to the limited range of the interrogating neutron flux within the MOX container.

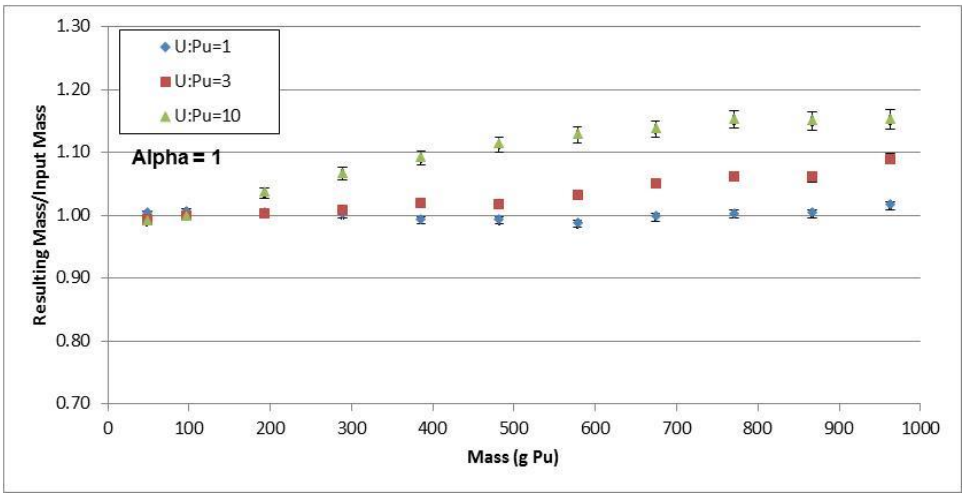

Fig. 17. Bias resulting from the Direct Measurement of Multiplication (DMM)/two-energy point model analysis integrated with a modified two energy point model for mixed oxide of various U:Pu ratios and with a fixed α (α =0.91). The plots show the ratio of the mass calculated from the modeled count rates to the plutonium mass input to the MCNPX simulation. The deviation between simulated measurement and the input value results is caused by the error in the DMM multiplication as shown in Fig. 18.

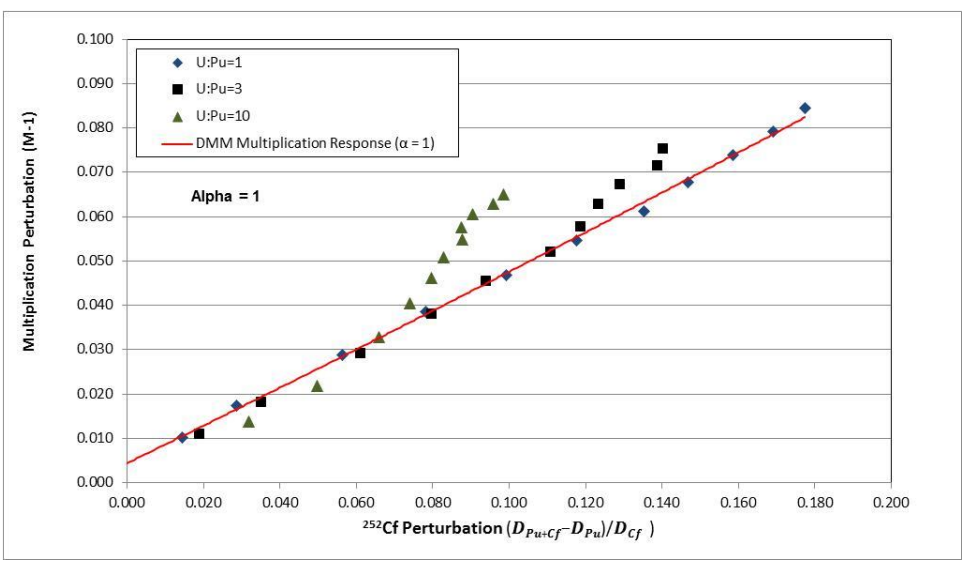

Fig. 18. The relationship between the sample multiplication and the active interrogation ratio for a series of mixed oxide masses modeled at three different U:Pu ratios. The multiplication determined by the Direct Measurement of Multiplication is also depicted in the plot.

A slight modification to the DMM calculation was necessary to include the effect of the U:Pu ratio.

$$\mathrm{M_{DMM}} = \mathrm{a}_\rho \cdot \left[\left(\frac{D_{Pu+Cf}-D_{Pu}}{D_{Cf}}\right) + \mathrm{r_{UPu}} \cdot \left(\frac{D_{Pu+Cf}-D_{Pu}}{D_{Cf}}\right)^{\mathrm{n}}\right] + \mathrm{b}_\rho \ , \tag{12}$$

where

D_{Pu+Cf}	=	the doubles rate with both ^{252}Cf and the MOX container in the cavity,
D_{Pu}	=	the doubles rate with only the MOX container in the cavity,
D_{Cf}	=	the ^{252}Cf doubles rate without the MOX container,
r_{UPu}	=	the U:Pu ratio (measured using MGA or other gamma-ray isotopic analysis),
a_ρ and b_ρ	=	empirically determined constants
n	=	empirically determined constant, from MCNPX n = 3 for this geometry.

This correction to the DMM measurement is purely empirically determined at this time. Additional work will be necessary to provide a physics-based rationale for this simple correction factor. A plot of the multiplication and interrogation ^{252}Cf perturbation values for a series of simulated samples containing between 50 and 1000 g of Pu and U:Pu ratios of 1, 3, and 10 is provided in Fig. 19, and the relative mass results are shown in Fig. 20.

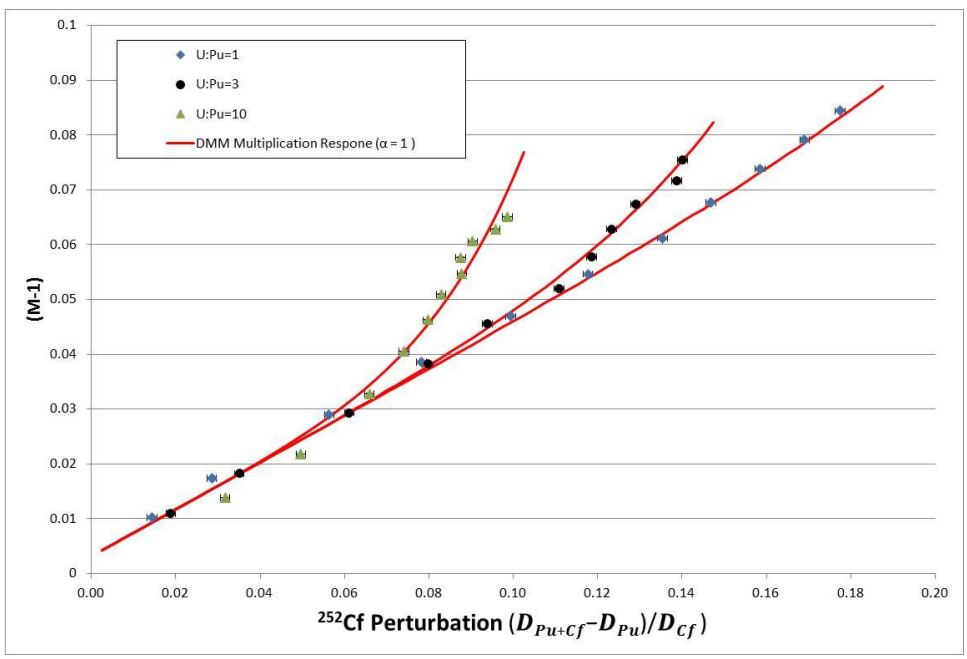

Fig. 19. The relationship between the sample multiplication and the active interrogation ratio for a series of mixed oxide masses modeled at three different U:Pu ratios. The multiplication determined by the modified Direct Measurement of Multiplication result is also depicted in the plot.

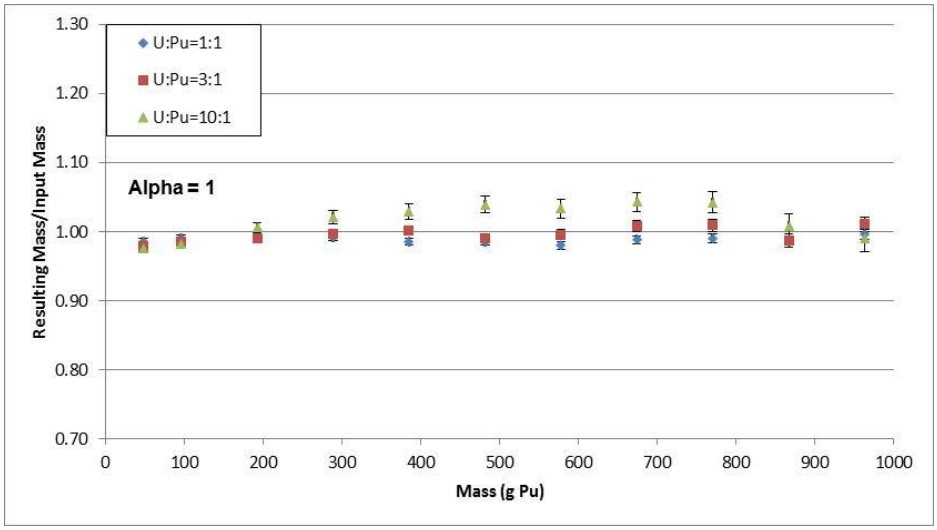

Fig. 20. Expected measurement performance of the Direct Measurement of Multiplication technique integrated with a modified Two Energy Point Model [5] for MOX with (α = 0.91) for various U:Pu ratios. The plots show the ratio of the mass calculated from the modeled count rates to the plutonium mass input to the MCNPX simulation. The deviation between the resulting simulated mass and the input value results is caused by a limitation of the point model. Note: Error bars are MCNPX precision values – not expected measurement precision.

7.4 HIGH ALPHA AND HIGH U:PU RATIO

Items with both high (α, n) and high U:Pu ratios are expected to be adequately treated by the combination of the 2 energy model discussed in Section 7.2 and the modified DMM algorithm shown in Eq.12. To verify this performance, we re-examine the same simulated item set discussed in Section 7.3. Figure 21 plots the same data shown in Fig. 19 however this time with **α=10** while the curves in red represent the DMM response function for **α=1**. The deviation between the DMM response curves and the item's multiplication is due to the difference the average neutron energy between the ^{240}Pu spontaneous fission neutrons and the (α, n) source and illustrates the importance of the 2 energy model for high (α, n) samples.

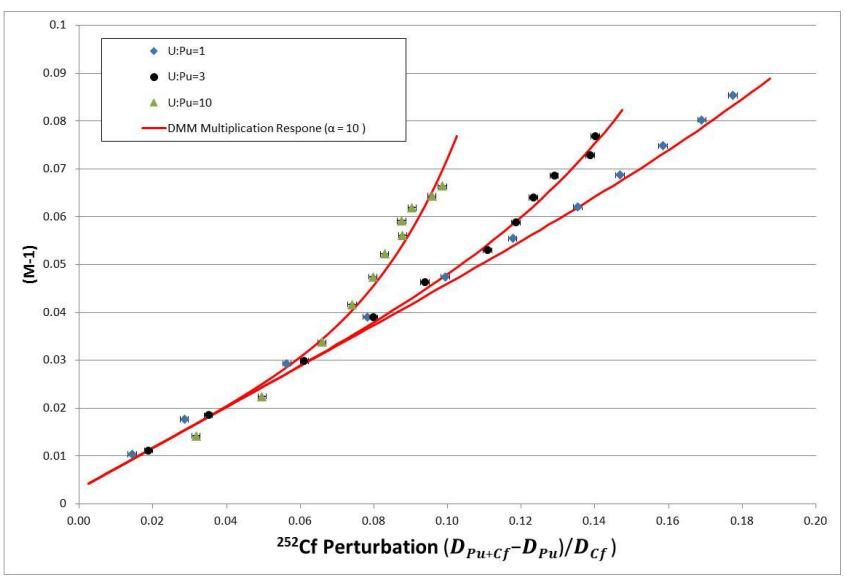

Fig. 21. The relationship between the sample multiplication and the active interrogation ratio for a series of mixed oxide masses modeled at three different U:Pu ratios for high alpha items. The multiplication determined by the modified Direct Measurement of Multiplication result is also depicted in the plot.

Figure 22 shows the expected measurement bias for MOX assay with U:Pu ratio of 10:1 using the simple DMM analysis but without the 2 energy model or U:Pu correction. Applying the 2 energy point model and UPu correction to these simulated measurements, we plot the ratio of simulated measurements versus the input mass values in Fig 23. Note that the scatter seen in this plot is due to the statistical limitations of the MCNPX simulations performed and do not reflect the expected measurement precision. The expected measurement precision is shown in Fig. 11 above. In Fig. 23 we see an apparent trend in the U:Pu=10:1 result. This corresponds to portion of the U:Pu=10:1 simulated measurement points where the curvature is opposite to the DMM response. Such trending could occur due to the neutron detection energy dependence of the HLNCC-II or more likely due to scattering from ^{238}U. However, even if this trending is present it is expected to be small relative to the predicted 5 to 10% precision expected for an α=10 sample.

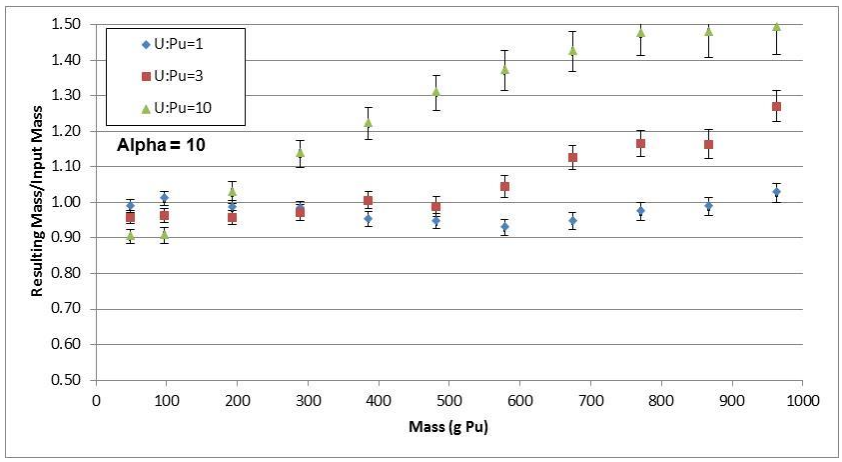

Fig. 22. Expected measurement performance of the Direct Measurement of Multiplication technique integrated with the traditional point model and without the U:PU ratio correction for MOX items with (α = 10) for various U:Pu ratios. The plots show the ratio of the mass calculated from the modeled count rates to the plutonium mass input to the MCNPX simulation. Note: Error bars are MCNPX precision values – not expected measurement precision.

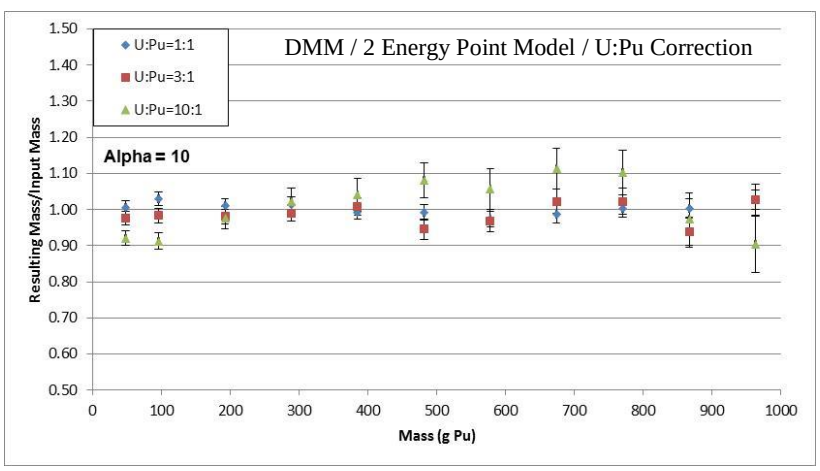

Fig. 23. Expected measurement performance of the Direct Measurement of Multiplication technique integrated with a modified Two Energy Point Model [5] for MOX with (α = 10) for various U:Pu ratios. The plots show the ratio of the mass calculated from the modeled count rates to the plutonium mass input to the MCNPX simulation. Note: Error bars are MCNPX precision values – not expected measurement precision.

7.5 DMM ACCURACY

Based on this initial analysis of the potential biases inherent in the DMM analysis, the overall accuracy of the DMM technique over a broad range of U:Pu ratios, alpha, and mass, is expected to be approximately 1 to 2%, depending on the quality of the base multiplication response calibration. However, potential biases arising from the specific chemical impurities in the MOX (e.g., fluorides) have not yet been examined.

8. CONCLUSIONS

A combination of scoping measurements and MCNPX calculations suggests that the DMM technique can offer significant performance improvement for the assay of plutonium oxide and MOX scrap materials. With this technique, a relatively modest neutron coincidence counter can provide a level of performance and throughput similar to the level achievable using a high-performance multiplicity counter. The primary benefit of this approach is the large reduction in the amount of ^{3}He required to achieve the same measurement results. For example, the HLNCC-II (employing the DMM technique) requires about *1/18* the ^{3}He needed to fabricate a high-end epi-thermal multiplicity counting system.

However, the DMM technique is an active interrogation technique. As is typical of active neutron interrogation measurements, the system must be calibrated with representative, well-characterized standards in a reproducible geometry and is susceptible to changes in source-sample coupling. That is, changes in the relative orientation of the ^{252}Cf interrogation source and the sample can introduce bias into the assay result.

The ^{252}Cf source strength used for the performance projections depicted in Fig. 11 may be increased to offer additional improvement in the measurement result. However, count rate limitations of the electronics, and uncertainty in the dead-time corrections, will ultimately limit the maximum practical interrogating source size. (Note: Unlike in the traditional AWCC measurement, a larger interrogating neutron source can improve the measurement precision.)

The potential biases seen in Fig. 23 can not be readily confirmed or eliminated through MCNP since these results are based on assumptions about the simulated measurement as well as the accuracy of the nuclear data within the MCNP structure. Instead these must be examined experimentally. However, some features can be controlled for instance, the HLNCC-II detection efficiency is neutron energy dependent due to its single ring of ^{3}He detector tubes. The non-linearity in response as in Fig. 15 is due in part to this energy dependence. Application of the DMM to multi-ring detector systems such as the AWCC or the boron straw based HLNCC will have a lesser dependence on neutron energy potentially resulting in a more linear response. This could be verified by repeating portions of this study for a multi-ring detector system.

Future Safeguards Technology Development Work

The results of this study indicate that the DMM technique is a viable approach to measurement of the self-multiplication of plutonium-bearing materials packaged within typical scrap and product MOX containers. The work suggests that a level of performance achieved by high-end multiplicity assay systems (requiring hundreds of liters of ^{3}He) could be achieved by a much less costly system such as the HLNCC (which requires only 20 liters of ^{3}He). The DMM technique could also be coupled with ^{10}B-lined systems currently under development at Los Alamos National Laboratory and Proportional Technologies, potentially eliminating the need for ^{3}He in these applications.

The next step in DMM development is to demonstrate the technique at an active fuel-processing facility. Since these facilities are generally bound by international safeguards protocols, the necessary neutron assay systems should already be available at the facilities. Only a reliable source positioning device and suitably sized interrogating ^{252}Cf source would need to be provided to adapt these systems for demonstration of the DMM technique. The greatest challenge appears to be access to a suitable fuel facility at which to conduct the demonstration.

This study has identified the need for a correction factor based on the U:Pu ratio of the MOX material. The empirical correction implemented for this study needs to be examined further to determine if it is suitable for use with real measurements. This could be accomplished by additional MCNP simulations or by measurement of a broad range of material forms.

Although the DMM technique is based on simple coincidence counting, as opposed to multiplicity analysis, the interpretation of the data is based on the point model and is susceptible to the same potential biases as the multiplicity counting systems. Development and vetting of advanced point-source models is required to minimize/eliminate these biases. Work by others on extending the point model has not yet been generally accepted or integrated into mainstream safeguards. The DMM technique not only would benefit from such modification but also would afford an ideal platform from which to evaluate these point model extensions.

REFERENCES

[1] R. D. McElroy, Jr. and S. Croft, "Preliminary Performance Results for a Direct Multiplication Measurement for Use in Neutron Multiplicity Analysis," in *International Nuclear Materials Management 55th Annual Meeting*, Atlanta, Ga, 2014.

[2] D. Davidson, R. D. McElroy, D. Brochu and M. Villani, "A New 208 Liter Drum Neutron Coincidence Counter with Add-A-Source Matrix Correction," in *Proceedings of the 36th Annual Meeting for the Insitute of Nuclear Materials Management*, Desert Springs, California, July, 1995.

[3] H. O. Menlove, J. Baca, W. Harker, K. E. Kronke, M. C. Miller, S. Takahashi, K. Kobayashi, S. Seki, K. Matsuyama and a. S. Kobayashi, "WDAS Operation Manual Including the Add-A-Source Function, LA-12292-M," Los Alamos National Laboratory, Los Alamos, NM, 1992.

[4] N. Ennslin and et.al., "Application Guide to Neutron Multiplicity Counting, LA-13422-M,," Los Alamos National Laboratory,, Los Alamos, NM, 1998.

[5] S. Croft, A. Favalli and R. D. McElroy, *Unpublished report,* Oak Ridge, TN, 2015.

[6] M. S. Krick, D. G. Langner and J. E. Stewart, "Energy-Dependent Bias in Plutonium Verification Measurements using Thermal Neutron Multiplicity Counters," Los Alamos National Laboratory document LA-UR-97-3427, Los Alamos, NM, 1997.

[7] H. Menlove, "Description and Operation Manual for the Active Well Coincidence Counter, Los Alamos National Laboratory report LA-7823-M," Los Alamos National Laboratory, Los Alamos , NM, 1979.

[8] P. Rinard, "Application Guide to Shufflers, LA-13819-MS," Los Alamos National Laboratory, Los Alamos, NM, (2001).

[9] J. S. Hendricks and et.al., "MCNPX Version 2.5, LA-UR-02-7086," Los Alamos National Laboratory, Los Alamos, NM, 2002.

[10] R. D. McElroy, Jr. and S. Croft, "The Role of Active Neutron Interrogation to Safeguards Non-Destructive Assay (NDA) in the Post He World," in *The 53rd Annual Meeting of the Institute of Nuclear Materials Management*, Orlando, FL, 2012.

[11] M. Tolbert, "New Brunswick Laboratory Certified Reference Material Certificate of Analysis," New

Brunswick Laboratory, Argonne, IL, 1999.

[12] Canberra Industries, "Model JCC-31 High Level Neutron Coincidence Counter Specification Sheet," [Online]. Available: http://www.canberra.com/products/waste_safeguard_systems/pdf/JCC-31-SS-C36906.pdf. [Accessed 31 March 2015].

[13] E. F. Shores, "SOURCES 4C: A Code for Calculating (,n), Spontaneous Fission,LA-UR-02-1839," Los Alamos National Laboratory, Los Alamos, NM, April 2002.

[14] G. Jacobs and H. Liskien, "Energy spectra of neutrons produced by α-particles in thick targets of light elements," *Ann. Nucl. Energy,* vol. 10, no. 10, pp. 541-552, 1983.

[15] S. Croft and P. Chard, "Neutron Characterization of Plutonium Oxide Reference Samples at Harwell," in *15th Annual Symposium on Safeguards and Nuclear Material Management*, Rome, Italy, 1993.