Alternatives to Truncated Multiplicity Analysis to Improve Verification of Uranium Fuel Cycle Materials

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Abstract. More accurate verification of $^{235}$U enrichment and mass in UF$_6$ storage cylinders and of UO$_2$F$_2$ holdup contained in process equipment is needed to improve international safeguards and nuclear material accountancy at uranium enrichment plants. Small UF$_6$ cylinders (1.5” and 5” diameter) are used to store the full range of enrichments from depleted to highly-enriched UF$_6$. For independent verification of these materials, it is essential that the $^{235}$U mass and enrichment measurements do not rely on facility operator declarations. Furthermore, in order to be deployed by IAEA inspectors to detect undeclared activities (e.g. during complementary access), it is also imperative that the measurement technique is quick, portable, and sensitive to a broad range of $^{235}$U masses. Truncated multiplicity analysis is a technique that reduces both the value and the variance of the measured neutron count rates by only considering values 0, 1, and 2 of the multiplicity distribution. This is especially important for reducing the uncertainty in the measured doubles and triples rates in environments with a high cosmic ray background relative to the uranium signal strength. However, we believe that truncated multiplicity analysis may throw away too much useful data by truncating the distribution at 2. This paper investigates two alternative methods to truncated multiplicity analysis: 1) uses a 1σ outlier rejection limit to truncate the multiplicity distribution based on the measured data and 2) uses a second identical detector next to the detector measuring the item to directly track the cosmic ray variation. Experimental measurements of small UF$_6$ cylinders and UO$_2$F$_2$ working reference materials were performed at Los Alamos National Laboratory (LANL). The data were analyzed using the alternative methods and traditional truncated multiplicity analysis to compare the impact of the methods on the measured count rates and uncertainties. The results from this analysis directly support nuclear safeguards at enrichment plants and provide a more accurate verification method for UF$_6$ cylinders and uranium holdup in high...
background environments.
1. Introduction

Safeguarding uranium enrichment technology is important to verifying that State’s are in compliance with their obligations under the Nuclear Nonproliferation Treaty (NPT). Accurate verification of $^{235}$U mass and enrichment in uranium hexafluoride (UF$_6$) storage cylinders as well as the uranyl fluoride (UO$_2$F$_2$) holdup contained in process equipment is needed to reliably implement safeguards and nuclear material accountancy at uranium enrichment plants. Due to the broad range in the amount of $^{235}$U mass that can be stored in small UF$_6$ cylinders, it is important that the measurement technique used for verification is sensitive to the bulk item and to $^{235}$U mass ranging from grams to kilograms $^{235}$U.

Truncated multiplicity analysis is commonly used for assay measurements of very small Pu and U items when the cosmic ray background interferes with the measurement. This is a common challenge when operating close to the limit of detection. The interaction of cosmic rays with the environment, materials in the detector, and also with the assay item is a source of background neutrons. Spallation interactions resemble fission in that several neutrons can be emitted and a cascade can form boosting the number. The singles, doubles and triples rates expressed in counts per sec per gram for a medium to heavy weight target with a molar mass, $A$, are expected to scale roughly as $A^{2/3}$, $A^{5/3}$, $A^{8/3}$ respectively \cite{1,2}. This is why when weak sources are to be measured it is important to limit the high-Z content materials in and around the neutron detector. External shielding can reduce the influence of room neutrons although care must be taken that it does not in turn act as a source of cosmic ray induced neutrons. Furthermore, because the cosmic ray background rate is not fixed but varies with solar activity and weather conditions, especially the atmospheric pressure, the background should be tracked over the course of a measurement campaign.

Truncated multiplicity analysis uses only the first three multiplicity bins (zeros, ones, and twos) in the multiplicity distributions and ignores the higher multiplicities. This can be especially important for reducing the uncertainty in the measured doubles and triples rates in environments with a high cosmic ray background relative to the uranium signal strength. However, we believe that truncated multiplicity analysis may throw away too much useful data by truncating the distribution too soon.

This paper investigates two alternative methods to truncated multiplicity analysis: 1) uses a 1σ outlier rejection limit to truncate the multiplicity distribution based on the measured data and 2) uses a second identical detector next to the detector measuring the item to directly track the cosmic ray variation. A series of measurements of small UF$_6$ cylinders and UO$_2$F$_2$ working reference materials (WRMs) were performed at LANL using a well counter called the Mini Epithermal Neutron Multiplicity Counter (mini-ENMC) \cite{3}. The experimental data were analyzed using the alternative methods and traditional truncated multiplicity analysis to compare the impact of the two methods on the measured count rates and uncertainties. The results from this analysis directly support nuclear safeguards at enrichment plants and can improve the accuracy of verification methods for UF$_6$ cylinders and uranium holdup in environments where the background is high relative to the neutron signal from uranium.

2. Uranium Standards and Experimental Setup

We performed a series of experimental measurements of small UF$_6$ cylinders and UO$_2$F$_2$ WRMs using the mini-ENMC, at two facilities at LANL. There are two identical mini-ENMCs available at LANL for nuclear safeguards measurements. The mini-ENMC consists of 104 $^3$He tubes at 10-atm pressure that are arranged in four concentric rings and embedded in high density polyethylene. The mini-ENMC has a high efficiency, $\varepsilon$, of ~62% for a $^{252}$Cf point source at the center of the empty cavity. The axial detector efficiency profile is quite uniform across the items to be measured and also reasonably insensitive to the originating spectrum of the neutrons. The die-away time, $\tau_3$
(characteristic of the 1-D Rossi-α distribution) is also short, at about 19.1-μs. This means that the correction for accidental coincidences in coincidence counting mode is small. The high efficiency and short die-away time of the mini-ENMC enables measurements to be sensitive to a broad range of $^{235}$U masses, ranging from grams to kilograms.

For the UF$_6$ measurements, three different 1S cylinders (Monel, 1.5” diameter) and one 5B cylinder (nickel, 5” diameter) were measured in an above ground laboratory at LANL. The $^{235}$U enrichment ranged from 0.2wt% (depleted) to 69.6wt% $^{235}$U. Due to the different valve heights on the 1S cylinders, all measurements were performed without the lid on the mini-ENMC. This helped to minimize systematic uncertainties in the measurements. We also measured several UO$_2$F$_2$ WRMs (aluminum, 1” diameter) that were fabricated at the Portsmouth Gaseous Diffusion Plant (PORTS). These measurements were performed in a basement laboratory to reduce the natural cosmic ray background. The purpose of the UO$_2$F$_2$ measurements was to determine the absolute yield of $(\alpha,n)$ neutrons from fluorine induced by $^{234}$U α-particles [4]. The physical properties and isotopics of the UO$_2$F$_2$ WRMs were well characterized and traceable to a nationally recognized reference base. Figure 1 shows the $^{235}$U mass and enrichment values for the UF$_6$ and UO$_2$F$_2$ standards used in the measurements.

<table>
<thead>
<tr>
<th>Cylinder Type</th>
<th>Enrichment [wt%]</th>
<th>Mass [grams]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$^{235}$U</td>
<td>UF$_6$</td>
</tr>
<tr>
<td>1S</td>
<td>3.00%</td>
<td>8.85</td>
</tr>
<tr>
<td>1S</td>
<td>18.15%</td>
<td>72.24</td>
</tr>
<tr>
<td>1S</td>
<td>69.58%</td>
<td>177.43</td>
</tr>
<tr>
<td>5B</td>
<td>0.20%</td>
<td>12.00</td>
</tr>
</tbody>
</table>

FIG. 1. $^{235}$U enrichment and mass values for uranium standards used in measurements.

The uranium standards were measured in three different configurations shown in Fig. 2. It should be noted that for configuration #2, high density polyethylene inserts were used to expand the diameter of the mini-ENMC sample cavity in order to accommodate the 5B UF$_6$ cylinder. All of the standards were measured for long count times ranging from ~6-12 hours.

<table>
<thead>
<tr>
<th>WRM ID</th>
<th>Enrichment [wt%]</th>
<th>Mass [grams]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$^{235}$U</td>
<td>UO$_2$F$_2$</td>
</tr>
<tr>
<td>02</td>
<td>4.9530%</td>
<td>8.3299</td>
</tr>
<tr>
<td>17</td>
<td>4.9530%</td>
<td>8.7736</td>
</tr>
<tr>
<td>18</td>
<td>4.9530%</td>
<td>8.8225</td>
</tr>
<tr>
<td>26</td>
<td>4.9525%</td>
<td>8.9003</td>
</tr>
</tbody>
</table>
For all measurements, the signal from the mini-ENMC was split between a List Mode Multiplicity Module (LMMM) [5] and a JSR-15 multiplicity shift register [6]. Both data acquisition systems are used to count and record the total number of pulses and the time correlation of coincidence pulses. The list mode data is advantageous because it enables us to post process the data and change the gate and/or cycle time if needed to improve the uncertainties on the multiplicity measurements. The JSR-15 was connected to the laptop and International Atomic Energy Agency (IAEA) Neutron Coincidence Counting (INCC) [7] software version 5.1.2 was used to set the operating parameters for mini-ENMC, as well as collect and archive the measurement data. It is also important to note that several background measurements were performed for each configuration throughout the experiment in order to accurately quantify the cosmic ray background. LANL is approximately 2,242 meters above sea level and the cosmic ray background is ~7 times higher than at sea level.

3. Alternative to Truncated Multiplicity Analysis

One of the alternative methods to truncated multiplicity analysis was investigated by changing the outlier rejection limit in INCC from $3\sigma$ to $1\sigma$. The count time per cycle was kept constant at 30 seconds. An outlier in a group of cycles is defined as a cycle whose doubles and/or triples rates are greater than or equal to the user-specified number of standard deviations from the weighted average of the doubles and triples rates. The weighted mean of the doubles rate and outlier test for the modified truncation method are calculated by:

$$
\bar{D}_{\text{set}} = \frac{\sum_{i=1}^{N_c} D_i}{\sum_{i=1}^{N_c} \frac{1}{\sigma^2(D_i)}}, \quad \left| \frac{D_i - \bar{D}_{\text{set}}}{\sigma(D_i)} \right| \geq 1\sigma
$$

where $D_i$ is the doubles rate from $i$th cycle and $N_c$ is the total number of cycles in the data set. The statistical rejection is applied iteratively until a self-consistent data set is obtained. At each pass when a cycle is discarded, the total number of cycles is correspondingly reduced. As a result, the total number of bins in the multiplicity histogram is reduced because the higher multiplicities due to cosmic ray interactions were rejected as outliers.

Compared to traditional truncated multiplicity analysis, this method is essentially a statistical filtering of the measured rate data on a cycle by cycle basis which results in a truncated multiplicity histogram from outlier rejection. It is important to note that truncated multiplicity analysis truncates the multiplicity histogram at two and as a result both the measured rates and variance are reduced. The reduction in the measured rates varies with material type and geometry requiring careful
Advantages of this method over truncated multiplicity analysis are that it doesn’t throw away nearly as much of the measured signal from the item and can be used for long measurement times to remove the cosmic ray background without requiring post-processing of the measurement data (being an option in INCC). The main disadvantage is that the overall reduction in the doubles and triples measurement uncertainties are on average less than truncated multiplicity analysis.

The percent decrease in the 1σ of the doubles background and net doubles uncertainty using the 1σ outlier method and truncated multiplicity analysis is graphically shown in Fig. 3 for measurements in configurations #1 UO$_2$F$_2$ WRMs, #2 1-UF$_6$ cylinder, and #3 3-UF$_6$ cylinders measurements, respectively. The effect of 1σ outlier method and truncated multiplicity analysis on the measured doubles rates and uncertainty is also shown in Fig. 3.

**FIG. 3.** Percent decrease in 1σ net doubles for configurations #1, #2, and #3.

Figure 4 shows the factor decrease in the net doubles rate versus $^{235}$U mass for all of the uranium standard measurements. Comparing the measurement results from all the uranium standards, the overall decrease and relative standard deviation (RSD) of both methods on the doubles rate and the uncertainty was as follows:

- **Average decrease in doubles rate:** ~1% (factor of 1.01, 3% RSD) using 1σ outlier method.
and \( \sim 34\% \) (factor of 1.52, 6\% RSD) using truncated multiplicity analysis,

**Average decrease in doubles uncertainty**: \( \sim 18.6\% \) using 1\( \sigma \) outlier method and \( \sim 28.8\% \) using truncated multiplicity analysis.

![Graph showing factor decrease in net doubles rate versus \(^{235}\text{U} \) mass for all \( \text{U} \) standard measurements.](https://example.com/graph.png)

**FIG. 4.** Factor decrease in net doubles rate versus \(^{235}\text{U} \) mass for all \( \text{U} \) standard measurements.

Another alternative to truncated multiplicity analysis is to use a second identical detector next to the detector measuring the item to directly track the cosmic ray variation. For the configuration \#1 \( \text{UO}_2\text{F}_2 \) WRM measurements, we expected the correlated cosmic ray background to be dominated by interactions in the body of the detector because the items are small and low mass. To confirm this hypothesis, the background with a lead silicate WRM in the sample cavity was constantly measured using mini-ENMC \#2 (Channel 28) while measurements of the \( \text{UO}_2\text{F}_2 \) WRMs were being performed during the day. At night, both mini-ENMC \#1 (Channel 31) and mini-ENMC \#2 (Channel 28) were used to measure the background. In order to compare the effect of the lead silicate on the doubles and triples cosmic ray background, the sample cavity was empty for all measurements with mini-ENMC \#1. Figure 5 shows the background ratio in Channel 28 / 31 for the doubles and triples rates versus measurement date where Channel 28 contains lead silicate WRM in mini-ENMC \#2 and Channel 31 from mini-ENMC \#1 is empty.

Referring to Fig. 5, we see that the doubles and triples background rates measured with mini-ENMC \#2 (Channel 28) were \( \sim 21\% \) and \( \sim 10\% \) higher, respectively, due to the Pb silicate present in the sample cavity acting as a cosmic-ray target. For all of the background measurements of \( \sim 6-12 \) hours, the standard deviation in the Channel 28/31 ratio was calculated to be 6.5\% and 29\% for the doubles and triples rates, respectively. These results show that even for measurements made in a basement laboratory the correlated cosmic ray background due to interactions in the detector and item is significant and should be corrected for. Using a second identical detector next to the measurement is an expensive but powerful way to directly track the cosmic ray variation. Although short term fluctuations are strong, as one can see from the measured data, the average trends are

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1 Ratio of measured doubles rate to 1\( \sigma \) outlier or truncated multiplicity doubles rate.
better than alternating item and background measurements unless a sequence of short item in item out counts are made.

**FIG. 5.** Measured background ratio in Channel 28 / 31 for the doubles and triples rates versus measurement date where Channel 28 contains lead silicate WRM and Channel 31 is empty.

### 4. Summary and Conclusions

We performed a series of experimental measurements of small $\text{UF}_6$ cylinders and $\text{UO}_2\text{F}_2$ WRMs using the mini-ENMC at two different facilities at LANL. The $^{235}\text{U}$ enrichment ranged from 0.2wt% (depleted) to 70wt% $^{235}\text{U}$ and the corresponding $^{235}\text{U}$ mass ranged from 8-g to 179-g$^{235}\text{U}$. Due to the high cosmic ray background at LANL from being located approximately 2,242 meters above sea level, the experimental results were analyzed using truncated multiplicity analysis and two alternative methods to compare the methods and their impact on the measured count rates and uncertainty. This is especially important when using passive neutron measurements for uranium assay because the spontaneous fission rate of uranium is several orders of magnitude lower than plutonium.

One alternative method to truncated multiplicity analysis was investigated by changing the outlier rejection limit in INCC from $3\sigma$ to $1\sigma$. Compared to truncated multiplicity analysis, this method is essentially a statistical filtering of the measured rate data on a cycle by cycle basis which results in a truncated multiplicity histogram from outlier rejection. Advantages of this technique over truncated multiplicity analysis are that it doesn’t throw away nearly as much of the measured signal and can be used for long measurement times to remove the cosmic ray background without requiring post-processing of the measurement data. However, the main disadvantage is that the overall reduction in the doubles and triples measurement uncertainties is on average less than the traditional truncated multiplicity analysis method. Comparing measurements of all the uranium standards, the doubles rate decreased on average by a factor of 1.01 using the $1\sigma$ outlier method and by a factor of 1.52 using truncated multiplicity analysis. The average percent decrease in the uncertainty in the doubles rate was $\sim$18.6% using $1\sigma$ outlier method and $\sim$28.8% using truncated multiplicity analysis. The results from this analysis directly support nuclear safeguards at enrichment plants and can improve the accuracy of verification methods for $\text{UF}_6$ cylinders and uranium holdup in high background environments. Future work will include an in depth comparison of MCNPX simulations to experimental measurements and analysis of the list mode data. From this, we will confirm and quantify the fraction of signal rejected along with the background and corresponding item specific calibration parameters appropriate.

### Acknowledgements
We would like to acknowledge the Department of Energy National Nuclear Security Administration (NNSA) Office of Nonproliferation and International Security for supporting this work.

References


