SAND2013-4657C

EXPERIMENTAL MEASUREMENT OF URANIUM HEXAFLUORIDE FAST NEUTRON SIGNATURES

Mark Gerling, Erik Brubaker, Scott Kiff, Laura Kogler, Peter Marleau, Stanley Mrowka, Wondwosen Mengesha, and Michael Streicher†
Sandia National Laboratories, Livermore, CA

ABSTRACT

Monitoring uranium enrichment remains an important problem for nonproliferation and safeguards. Current technologies which measure the ^{235}U enrichment in UF_6 cylinders require controlled conditions for accurate measurements, and utilize gamma and low energy neutron signatures which measure only the outside surface of large, dense UF_6 volumes. This results in a non-robust measurement which could be exploited through diversion. Fast neutron spectrometry and imaging can be applied to fill this gap, as the high energy neutrons are deeply penetrating and carry the potential to allow for whole cylinder measurements. While this measurement concept has been explored before in simulation, here we will discuss a practical deployment of the technique. A suite of detectors were deployed to the Paducah Gaseous Diffusion Plant in July of 2012, including several different liquid scintillator cells (EJ-309), a ^{3}He tube, and a large sodium iodide scintillator. We will focus on the fast neutron response of the liquid scintillator, our efforts to characterize multiple UF_6 30B cylinders of varying enrichment using spectral and rate information, and our attempts to compare our results to previous simulations. Initial efforts demonstrate the ability to correlate the shape of the neutron energy spectrum data to the enrichment of the 30B cylinder.

INTRODUCTION

The primary limitation of current UF_6 assay methods is the volume they are able to reliably evaluate. Current methods of assay include gamma measurements as well as thermal neutron counting [1]. Passive gamma spectroscopy is less precise and measures a larger area, but is still confined to the surface layer of the cylinder. Similar to gammas, thermal neutrons have a short path length through UF_6 and are largely insensitive to the material in the center of a cylinder. In addition, both gamma and thermal neutron methods are sensitive to perturbations in geometry. A more robust solution is desirable for international safeguards, where coordinated state-level diversion scenarios may be of concern.

The high energy neutron signature from UF_6 cylinders utilizes the high penetration of several MeV neutrons to probe further into UF_6 cylinders than other methods allow. Sandia has been investigating this approach to determine how accurate, robust and sensitive a neutron spectrometer can be for this application. The significant processes which enable neutron spectrometry assay are spontaneous fission in ^{238}U (distribution tail up to 10 MeV) and the alpha on fluorine reaction which produces neutrons up to 2.5 MeV. There will be a correlation between the ^{238}U mass and the number of neutrons detected above 2.5 MeV, while the ^{234}U will

†Michael Streicher is now with the University of Michigan.
Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

SAND #: 2013-???

scale with the neutron population below 2.5 MeV. It has been reported that ^{235}U is proportional to ^{234}U in mass based enrichment processes, which would allow us to indirectly measure ^{235}U.

While simulation results have shown promise for this method, this paper will focus on an exploratory experimental setup which directly measured UF_6 cylinders using a variety of liquid scintillator (EJ-309) cells.

EXPERIMENT

Liquid scintillator was chosen because of its decent energy resolution and ability to discriminate between gammas and neutrons via a pulse shape discrimination (PSD) parameter. This has proven a successful combination in the past for the Neutron Scatter Camera [3], a detector which was evaluated for potential applications here in UF_6 assay. Because two of the larger uncertainties in this method are the amount of information available in the neutron energy spectrum, as well as the gamma background, the initial deployment contained only a small collection of individual cells of varying size. Included were 2" x 2", 3" x 3", 2" x 5" and 5" x 5" right cylinder cells (depth by diameter) read out by 2" and 5" Hamamatsu photo-electron tubes (PMT).

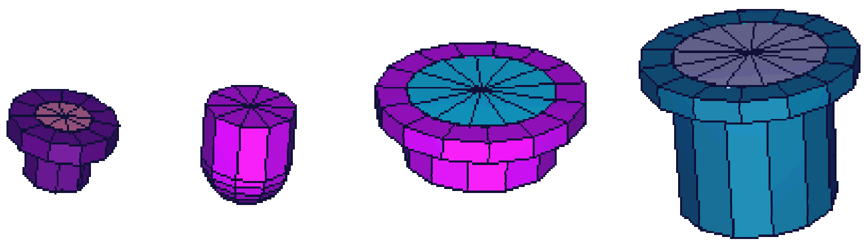

Figure 1. MCNP models of the 4 liquid scintillator cells, from smallest (2"x2") on left to largest (5"x5") on the right.

These detectors were arranged in a close packed configuration and packed in Styrofoam and cardboard for robust shipping in a large crate. Signal and power were run out of the crate and to an electronics rack which utilized a Mesytec MPD-4 module to read out energy and perform pulse-shape discrimination.

A total of 7 cylinders were measured by this setup, ranging from 0.72% to 4.95% enrichment, including two which were Russian in origin and reprocessed. We positioned our detectors between 0" and 40" from the cylinders, in side-on and end-on configurations. For the purpose of this comparison we will focus on ~40" distances in the side-on configuration where we had the most data for comparison. Lead blankets were also used in most cases for a degree of gamma shielding.

ID	Enrichment	Origin	Measurement Time	Distance
LU1511	0.72%	US	6 hours	10 inches
LU1603	2.00%	US	6 hours	38 inches
LU2081	3.60%	US	6 hours	38 inches
LU2550	4.95%	US	2.5 hours	38 inches
LU0296	4.00%	Russian	1.5 hours	38 inches
LU1004	4.95%	Russian	2 hours	39 inches

Table 1. Paducah 30B cylinder enrichments.

RESULTS

Data recorded for each cylinder consisted of energy and PSD for each detected event in list mode, allowing us to post-process the data and filter for neutron events before creating energy spectra. One common representation of this data is in an energy vs. PSD plot, which shows the neutron and gamma distributions. An example of this data for one of the cylinders is included below in figure 2.

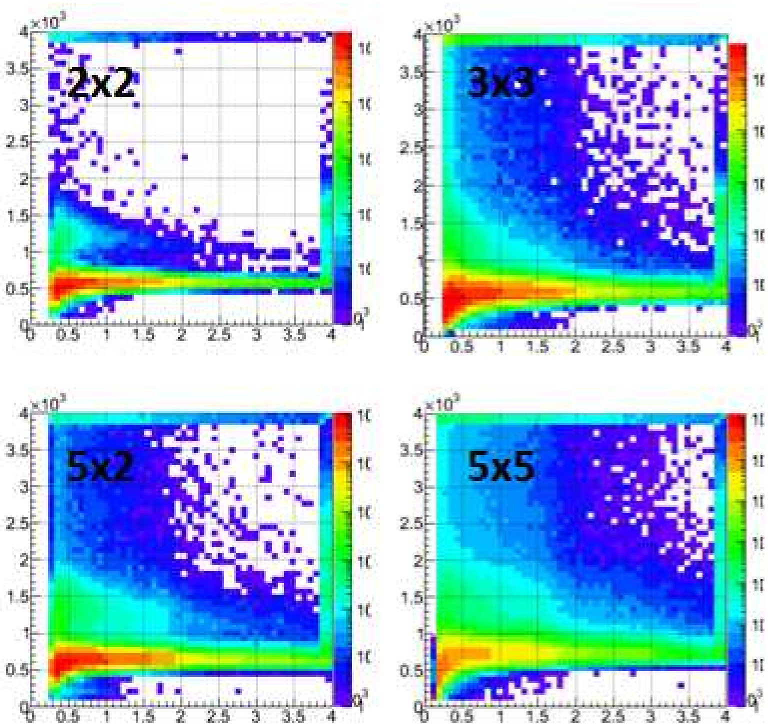

Figure 2. PSD vs. Energy. Units are in amplitude (arbitrary units). The 2” x 2” cell can be seen to have the best PSD separation.

The high gamma flux posed an issue for these measurements as pileup will negatively influence PSD measurements. This effect is apparent when performing a PSD cut for neutrons and plotting the energy spectrum (figure 3). Because of this, the larger cells suffered greatly in their ability to discriminate between gammas and neutrons, leaving the smallest 2” x 2” cell as the cleanest source of data. For this reason we focused further analysis on the 2” cell.

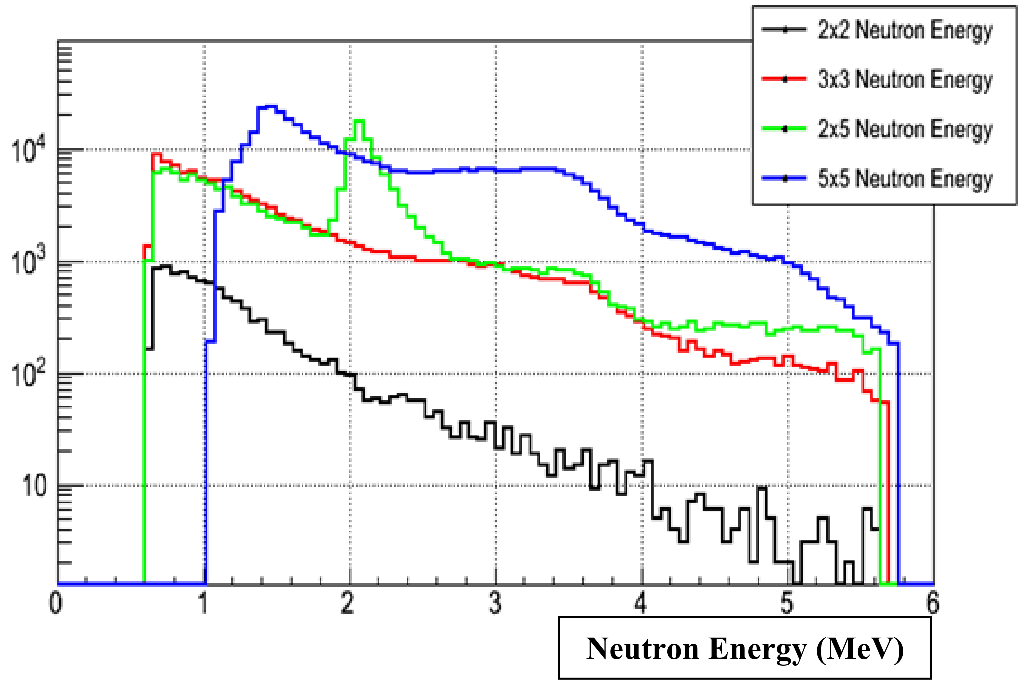

Figure 3. Neutron energy spectra after performing a neutron PSD cut, in units of MeV. A relatively smooth distribution is expected due to the poor energy resolution of liquid scintillator. Deviations from this can be attributed to gamma contamination.

Focusing in on the 2" x 2" cell allows us to plot a relatively simple energy spectrum comparison of the different cylinder enrichments shown in figure 4. It should be noted that the lowest enriched cylinder (black line) has the lowest low-energy component and the most high energy events, and the highest enrichment (pink line) has the largest low energy peak and fewer events in the high energy tail.

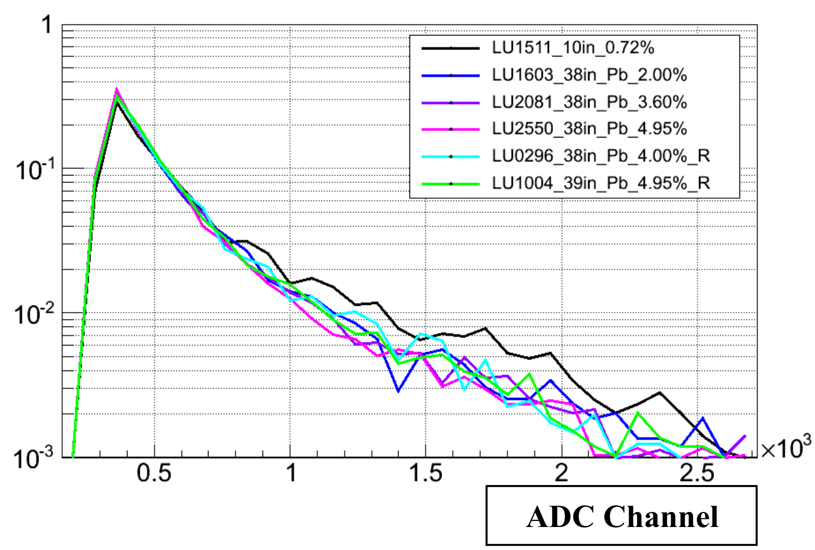

Figure 4. Normalized neutron energy spectra for 2" x 2" cell, all cylinders.

ANALYSIS
Due to the fact that the ^{234}U enrichment increases the neutron population under 2.5 MeV, and the de-enrichment of ^{238}U decreases the >2.5 MeV energy population, initial analysis focuses in on a two energy window ratio method. To avoid a biased result, optimization of this window was initially done using half of the available data up to a maximum of 3 hours. This optimized window fit was then used to correct the other half of the data (up to 3 hours again), and test the fit. The optimization looked at non-overlapping windows at least a few energy bins wide, and looped through many of these possible combinations while the 4 U.S. cylinder data sets were being fit to a linear function and tested for goodness of fit. The ideal fit was selected to have both a large slope and a small χ^2 value. Figure 5 contains the optimized window results for the first half data set. The fit result was then applied to the second half-data set, shown in figure 6.

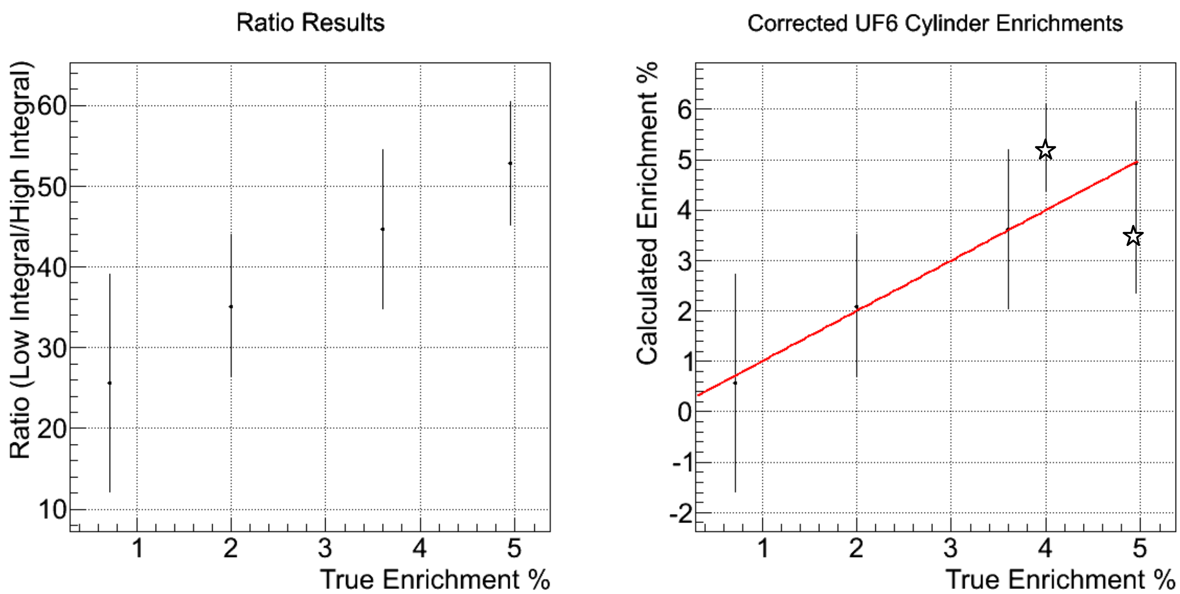

Figure 5. First half-data set with optimized windows. On the left are the ratio results vs. true enrichment. On the right are the corrected values using the inverse of the linear fit, with the Russian down-blended cylinders shown as well (denoted by stars).

The linear fit here resulted in a slope of 6.24, offset of 22.07, and Chi-squared of 0.01043 (not including the Russian down-blended). The prevailing error was due to the high energy integral statistics, where 100 high energy neutrons were counted on average. For the following result using the second half-data set, the same fit and windows were used, and the resulting Chi-squared was 1.3255. The typical uncertainty in these measurements was around 1.5% enrichment.

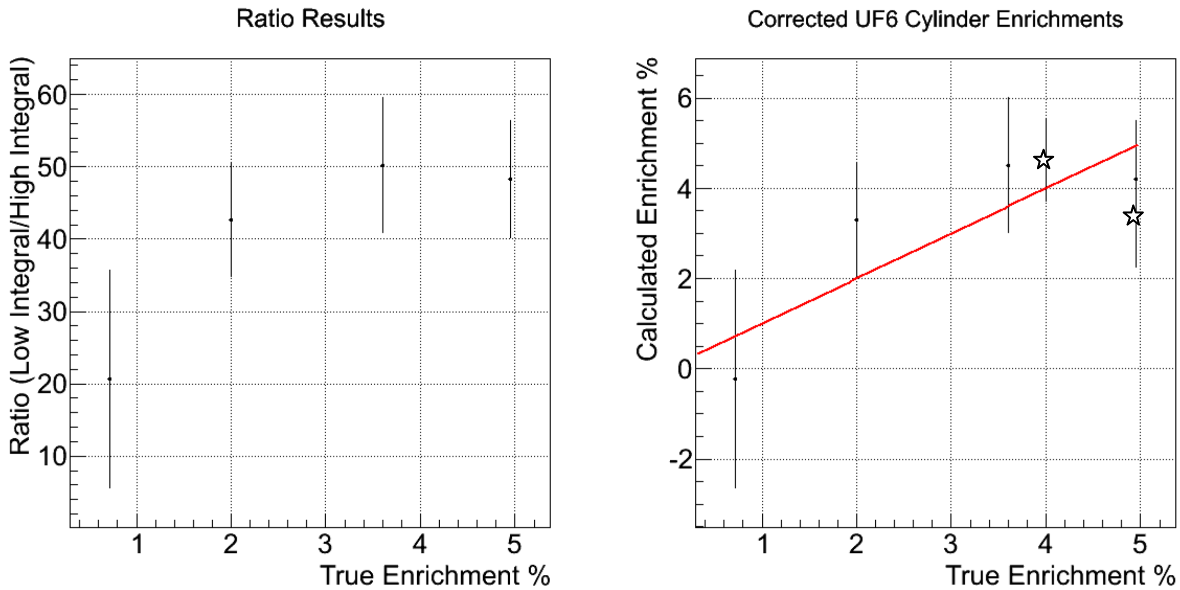

Figure 6. Second half-data set with first set optimized windows. On the left are the ratio results vs. true enrichment. On the right are the corrected values using the inverse of the linear fit, with the Russian down-blended cylinders shown as well (denoted by stars).

CONCLUSION

With this series of measurements we attempted to show the viability of performing uranium enrichment measurements using neutron spectrometry. While only a small set of cylinders were used in this study, this method holds promise as a viable technique, and further study is warranted. In addition, the other detector results may hold valuable data that can be analyzed with more time. Because these results represent a statistically limited measurement often having less than 100 events in the high energy integral, future measurements are desirable with a larger array of detectors. In future work we also hope to apply these methods and techniques to simulated data to improve our ability to simulate these complicated environments.

REFERENCES

1. [1] R. Berndt, E. Franke, P. Mortreau, Nucl. Instr. and Meth. A (2010), doi: 10.1016/j.nima.2009.10.060
2. [2] E.K. Mace, L.E. Smith, Nucl. Instr. and Meth. A (2010), doi:10.1016/j.nima.2010.09.149
3. [3] N. Mascarenhas *et al.*, “A measurement of the flux, angular distribution and energy spectra of cosmic ray induced neutrons at fission energies,” 2007 IEEE Nucl. Sci. Symp. Conf. Record, pp. 2050-2052.