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Experimental Measurement of Uranium Hexafluoride Enrichment using Fast Neutron Spectroscopy

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INTRODUCTION

Safeguards activities at uranium enrichment facilities require accurate, independent measurements of uranium mass for each UF_6 cylinder with contents that are either a process input or output. Current technology tends to rely upon gamma measurements (186 keV from ^{235}U decay) or thermal neutron counting using ^{3}He detectors [1]. Both methods are sensitive to geometrical perturbations of the UF_6 within the cylinder, giving rise to potentially significant systematic measurement uncertainties; also, given the short path length of low-energy gammas and neutrons through UF_6, these methods tend to be insensitive to material in the center of the cylinder. In the context of international safeguards, it is undesirable to rely upon a measurement technique that is effectively a measurement of the outer material skin; a full-volume assay is important for material balance at an enrichment facility.

Sandia is investigating the use of fast neutron spectrometers to augment the measurement capabilities surrounding UF_6 cylinder verification. Neutron spectrometry will allow for direct calculation of total mass and enrichment using two processes that generate significant neutron populations in the UF_6 [1]: spontaneous fission in ^{238}U (neutrons extending to about 10 MeV) and neutrons produced by bombardment of ^{234}U and ^{238}U decay αs upon fluorine, which terminate at about 2.5 MeV. ^{238}U mass will be proportional to the number of neutrons detected from spontaneous fission above 2.5 MeV; ^{235}U mass is reported in the literature to be proportional to ^{234}U in mass-based enrichment processes, and is determined by the neutrons counted below 2.5 MeV (total counts below 2.5 MeV minus the calculated ^{238}U spontaneous fission contribution and minus the corresponding ^{238}U (α,n) rate).

Although the processes that generate neutrons in UF_6 samples are readily acknowledged [2], to the best of the authors' knowledge neutron spectrometry has not been directly applied as a tool to independently measure ^{238}U and ^{235}U content of 30B cylinders. This technique has two strong advantages: first, it relies upon the measurement of highly-penetrating particles, making it possible to sample the entire UF_6 volume; second, using neutron spectrometry to determine the spontaneous fission and (α,n) components provides an independent measurement enrichment and total mass. Related publications [3] provide the foundation for this work. This summary discusses the first known enrichment measurement of UF_6 using neutron spectrometry.

DESCRIPTION OF THE ACTUAL WORK

Experimental Setup

Liquid scintillator was chosen because of its high detection efficiency, 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 Sandia's Neutron Scatter Camera [4], a detector which was evaluated for potential application to this enrichment measurement. 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 5.08 cm × 5.08 cm, 7.62 cm × 7.62 cm, 5.08 cm × 12.7 cm and 12.7 cm × 12.7 cm right cylinder cells (depth by diameter) read out by either a 5.08 cm or a 12.7 cm Hamamatsu photomultiplier tubes (PMT).

Fig. 1. MCNP models of the 4 liquid scintillator cells, rotating clockwise from smallest (5.08 cm × 5.08 cm) on top-left to largest (12.7 cm × 12.7 cm) on the bottom-left.

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.

The measurements were performed at the Paducah Gaseous Diffusion Plant near Paducah, KY. An aerial image depicting the measurement location is shown in Fig. 2. A total of seven 30B cylinders were measured by this setup, ranging from 0.72% to 4.95% ^{235}U enrichment, including two of Russian origin containing material down-blended from HEU. We positioned our detectors up to 1 meter from the cylinders, in side-on and end-on configurations. For the purpose of this comparison we will focus on 1 meter 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.

Fig. 2. The measurement area (red box) is shown in relation to a source of background (yellow box); other cylinders within the blue box are new, unfilled cylinders and thus do not contribute to the neutron background.

TABLE I. Paducah 30B cylinder enrichments.

ID	Enrichment	Origin	Dwell Time	Distance
1	0.72%	US	6 h	25 cm
2	2.00%	US	6 h	1 m
3	3.60%	US	6 h	1 m
4	4.95%	US	2.5 h	1 m
5	4.00%	Russian	1.5 h	1 m
6	4.95%	Russian	2 h	1 m

Measured Data

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 a PSD vs. energy plot, which shows the neutron and gamma distributions. An example of this data for one of the cylinders is included below in Fig. 3.

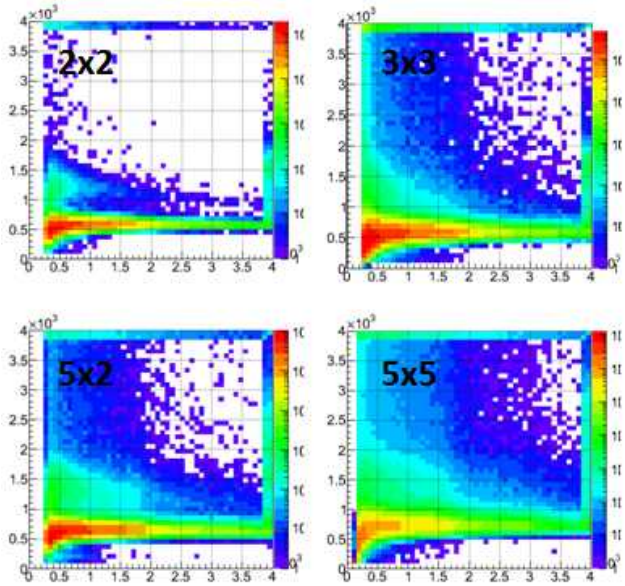

Fig. 3. PSD vs. energy for each EJ-309 cell. Units are in amplitude (arbitrary units). The horizontal band near PSD=0.5 represents gamma rays; neutrons populate a horizontal region between PSD=[1.0, 1.5]. The 5.08 cm × 5.08 cm cell can be seen to have the best PSD separation.

The high gamma flux posed an issue for these measurements as pileup will negatively influence PSD. This effect is apparent when performing a PSD cut for neutrons and plotting the energy spectrum (Fig. 4). Because of this, the larger cells suffered greatly in their ability to discriminate between gammas and neutrons, leaving the smallest 5.08 cm × 5.08 cm cell as the cleanest source of data. For this reason we focused further analysis on the 5.08 cm × 5.08 cm cell.

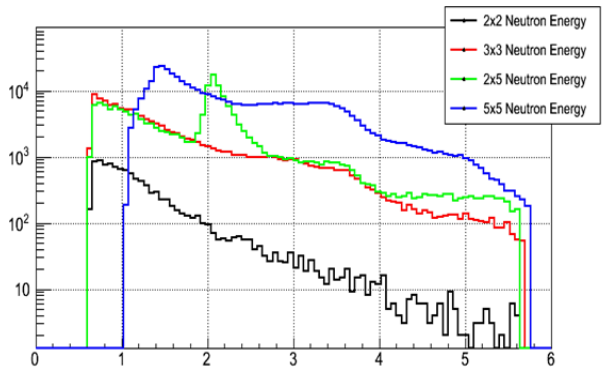

Fig. 4. Neutron energy spectra after performing a neutron PSD cut, in energy units of MeV. A relatively smooth distribution is expected due to the poor energy resolution of liquid scintillator. Deviations from this can be attributed to gamma contamination. The plot legend relies upon a naming convention using cell dimensions given in inches.

Focusing on the 5.08 cm × 5.08 cm cell allows us to plot a relatively simple energy spectrum comparison of the different cylinder enrichments shown in Fig. 5. It should be noted that the cylinder with naturally-enriched material (black line) has the smallest 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. Poor statistics at high energies may cast doubt on this assertion, but re-binning the high-energy region into fewer bins with more counts in each bin provides confidence in this statement.

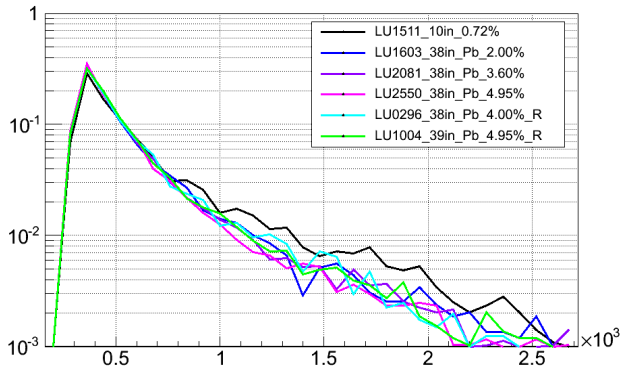

Fig. 5. Normalized neutron energy spectra for 5.08 cm × 5.08 cm cell, all cylinders.

RESULTS AND DISCUSSION

Analysis

Due to the fact that the ^{234}U enrichment increases the neutron population under 2.5 MeV, and the depletion of ^{238}U decreases the neutron population above 2.5 MeV, initial analysis focuses in on a two energy window ratio method. The ratio method has been trained on the first half of each data set (Fig. 6) and then applied to the second half of each data set (Fig. 7). This provides a crude sense of the reproducibility of each measurement.

The optimization of the ratio test used the restriction of non-overlapping windows at least a few energy bins wide; this restriction was enforced because the statistics in any one energy bin are poor at high energies, and allowing the test metric to consider a single bin may converge on a fit with poor uncertainty. The ideal fit was selected to have both a large slope and a small χ^2 value.

The test metric optimization considered only the four non-Russian cylinder data sets, although the metric was then applied to the Russian down-blended material measurements to determine how the neutron spectrum of these cylinders compares. Fig. 6 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 Fig. 7.

The linear fit here resulted in a slope of 6.24, offset of 22.07, and χ^2=0.01043 (not including the Russian down-blended material). 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 χ^2 was 1.3255. The typical uncertainty in these measurements due to counting uncertainty corresponded to 1.5% ^{235}U enrichment.

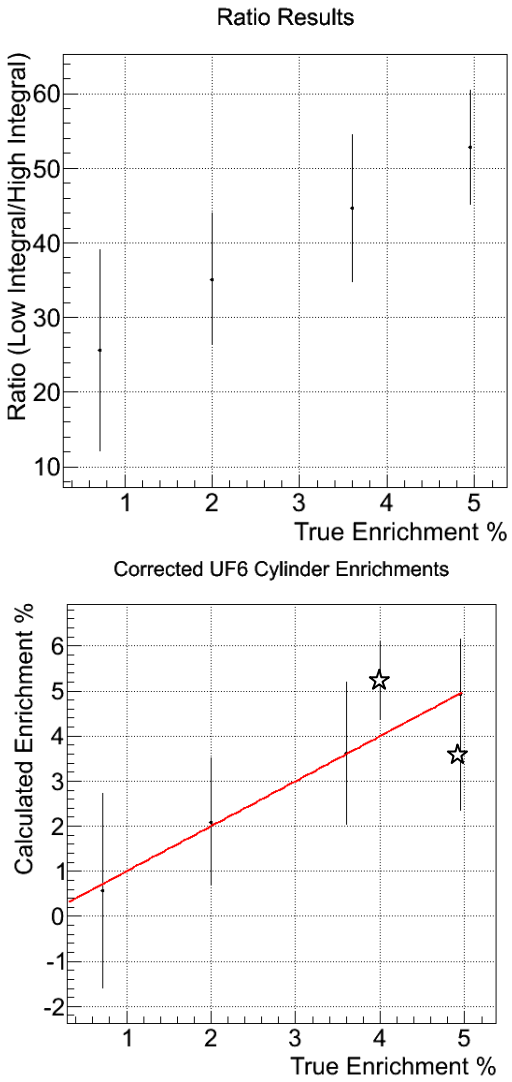

Fig. 6. First half-data set with optimized windows. On the top are the ratio results vs. true enrichment. On the bottom are the corrected values using the inverse of the linear fit, with the Russian down-blended cylinders shown as well (designated by stars).

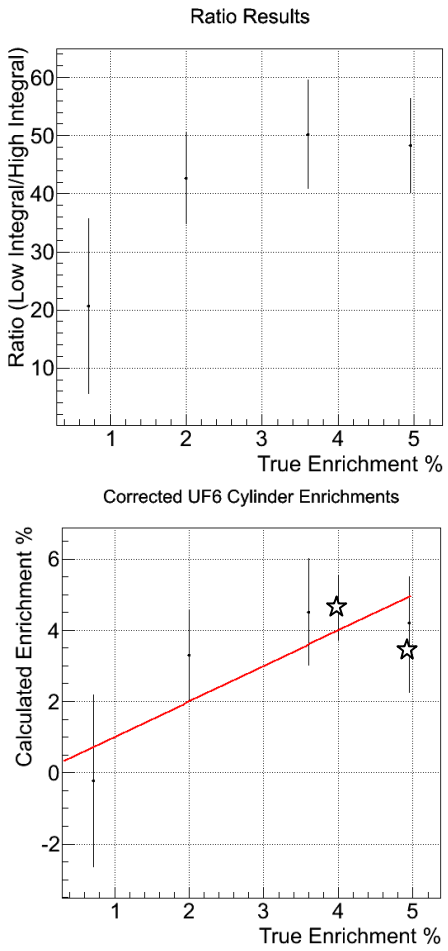

Fig. 7. Second half-data set with first set optimized windows. On the top are the ratio results vs. true enrichment. On the bottom are the corrected values using the inverse of the linear fit, with the Russian down-blended cylinders shown as well (designated by stars).

Conclusion

This series of measurements was performed in an attempt to show the viability of performing uranium enrichment measurements using neutron spectrometry. While only a small set of cylinders were used in this study, the results confirm that neutron spectroscopy can indeed be used to measure enrichment of UF_6.

Comparison of Fig. 6 and Fig. 7 indicates the neutron energy spectrum remained reasonably stable over a multiple-hour outdoor measurement, as the ratio test optimized on the data of Fig. 6 also fits the data of Fig. 7 within statistical uncertainty. The Russian down-blended material does not conform to this linear relationship, but it was not necessarily expected to have the same neutron energy spectrum, as the isotopic content of ^{234}U is different in this material due to the initial enrichment to HEU with subsequent down-blending to LEU.

Further analysis of this data may reveal valuable information from the larger scintillator cells that can be extracted in time. Because the results obtained with this small detector array represent a statistically limited measurement often having fewer 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.

NOMENCLATURE

EJ-309 = Eljen Technology EJ-309 liquid scintillator
HEU = highly-enriched uranium
LEU = low-enriched uranium
PMT = photomultiplier tube
PSD = pulse shape discrimination

ENDNOTES

[1]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.

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