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# FAST NEUTRON SIGNATURES FOR URANIUM HEXAFLUORIDE ENRICHMENT MEASUREMENTS

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## ABSTRACT

One of the most important problems in nonproliferation is monitoring the degree of enrichment from uranium enrichment plants. Centrifuge enrichment plants are necessary to produce fuel for commercial power plants but are also capable of producing highly enriched uranium, which can be configured into a nuclear weapon. Existing technologies for measuring $^{235}U$ enrichment in a $UF_6$ container (for example, a 30B cylinder) require controlled conditions for accurate measurements, making them susceptible to diversion scenarios. Low-energy neutrons and gammas used by current technologies have short penetration through dense $UF_6$, and since the $UF_6$ thickness near the measurement location is not known *a priori*, the current measurement techniques are insensitive to $UF_6$ in the center of the detector. Fast neutron spectrometry and imaging can be applied to ascertain the $UF_6$ enrichment inside the cylinder. Deeply-penetrating fast neutrons allow measurements that are sensitive to materials throughout the entire cylinder. Imaging the $UF_6$ material distribution within the cylinder allows compensation of geometry-dependent measurements when the $UF_6$ mass is unevenly distributed. The measurement concept, applications, limitations, and simulation results are presented in this submission.

## 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 (see Figure 1). In the context of international safeguards, for which coordinated state-level diversion scenarios are a concern, it is undesirable to rely upon a measurement technique that is effectively a measurement of the outer material skin, blind to the central contents.

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 $\alpha$s upon fluorine, which terminate at about 2.5 MeV. $^{238}U$ mass will be proportional to the number of neutrons detected from

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spontaneous fission above 2.5 MeV; $^{235}$U mass is proportional to $^{234}$U, 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 ($\alpha$,n) rate). Figure 2 shows the contributions to the $UF_6$ neutron spectrum.

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 ($\alpha$,n) components provides an independent measurement enrichment and total mass.

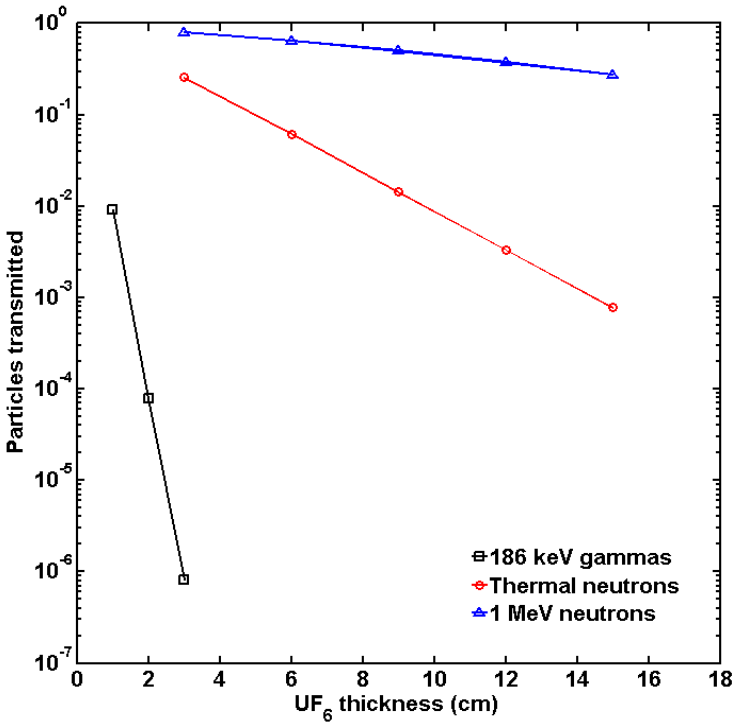


**Figure 1. A Monte Carlo calculation of the particles transmitted through a given thickness of $UF_6$ up to a maximum thickness expected for a uniform material loading in a 30B cylinder. All calculations consider a beam of incident particles normal to a slab of $UF_6$. Neutron measurements do not consider the effects of induced fission. High-energy neutrons have far superior sensitivity to material in the center of a cylinder.**

There is much work to be accomplished in applying spectrometry to $UF_6$ cylinder enrichment measurements. First, the neutron emissions must be accurately simulated to understand how scattering and self-absorption in the $UF_6$ modify the energy spectrum, which will likely make spectrometry more challenging. It is also important to study detector materials and configurations to optimize the system for spectrometry, as these parameters can affect overall neutron sensitivity and energy resolution. Laboratory experiments will be needed to validate the simulations, and a field test is desired to gauge overall system performance by measuring $UF_6$ cylinders at an enrichment or fuel fabrication facility. By the end of this project, the authors expect to produce an instrument that is sensitive to $UF_6$ material throughout a cylinder and that can be used to verify $UF_6$ cylinders in

storage with measurement times on the order tens of minutes to a few hours, depending upon the ability to optimize the measurement technique.

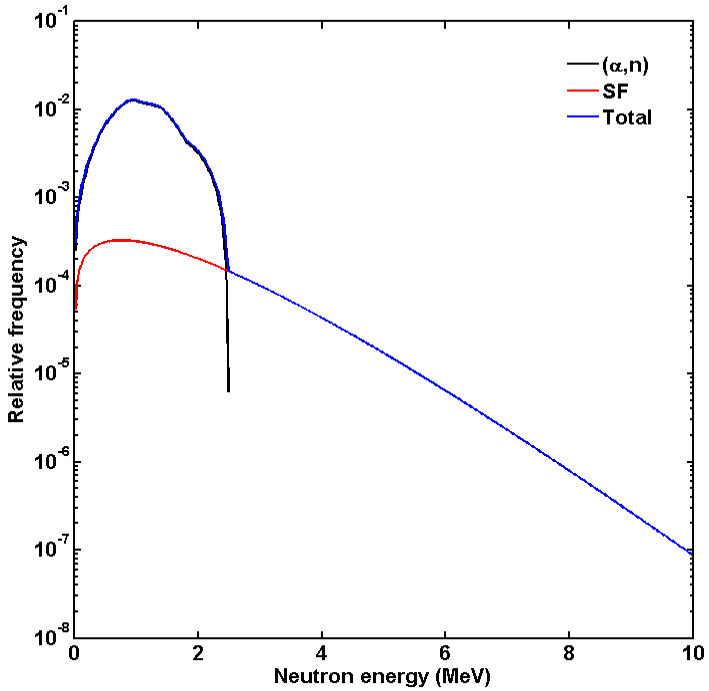


**Figure 2. A plot of the neutron spectral components for 5% enriched $UF_6$. The $^{234}U$ (α,n) reaction on fluorine generates a peaked neutron spectrum that terminates abruptly near 2.5 MeV. The $^{238}U$ spontaneous fission spectrum peaks near this upper boundary, and extends out to about 10 MeV. Placing an energy cut at 2.5 MeV will allow separation of the two contributions to the neutron source term.**

## SIMULATIONS

The principal goal of this project is to take advantage of changes in the neutron energy spectrum arising from the degree of uranium enrichment. Therefore, before even considering the detector response function, it is useful to study the pure neutron spectrum emitted from the cylinder, and how the original spectrum is altered by scattering, attenuation, and induced fission in the cylinder. It is possible that the differences in the spectrum can be washed out by these processes, making the neutron spectrometry technique unusable in practice—although from initial calculations it does appear that the spectral differences remain sufficient for neutron spectrometry to continue to be pursued as a viable enrichment measurement technique.

The neutron emission spectra calculated for this study use the isotopics presented in Berndt [1] for depleted uranium hexafluoride (DU), natural uranium ($^{nat}U$), and uranium enriched to 5% $^{235}U$ (LEU). SOURCES 4C [3] is used to calculate the spontaneous fission and (α,n) spectral contributions. The SOURCES 4C calculated spectral contributions are presented in Figure 3.

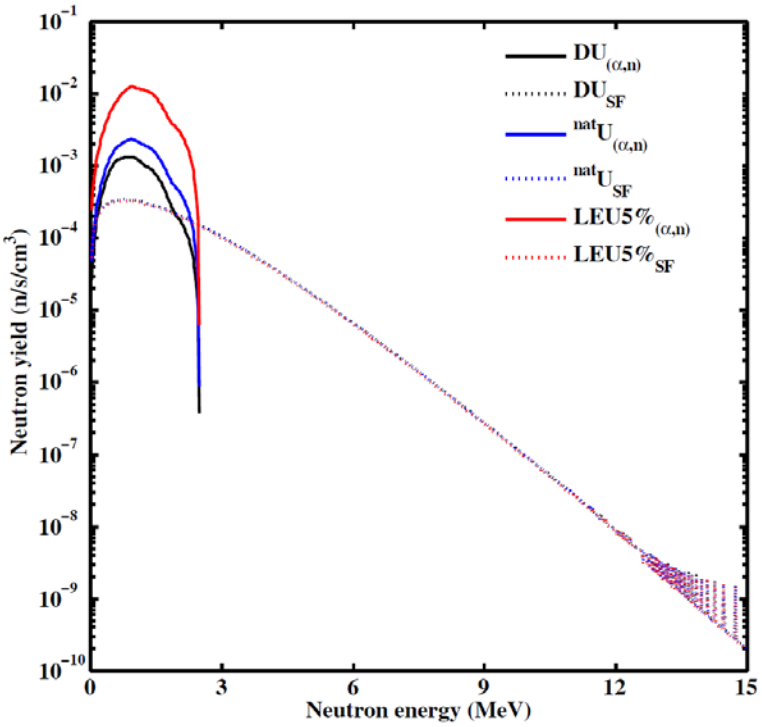


**Figure 3. The (α,n) and spontaneous fission (SF) contributions to the neutron energy spectrum for three enrichment levels of $UF_6$, plotted separately. The spontaneous fission spectral contributions are nearly identical for each enrichment, since the total $^{238}U$ mass is approximately constant. However, the (α,n) contribution varies by over an order of magnitude, changing the total energy spectrum in a quantifiable manner.**

To calculate the degradation of the energy spectra via scattering, capture, and induced fission in the $UF_6$, the SOURCES 4C spectra are inserted into an MCNP5 [4] model of a 30B cylinder [5]. This model considers a maximum loading of $UF_6$ with a uniform distribution of material in a thick shell. Since the initial measurement instrument is likely to have 12.7-cm right cylinders as detection volumes and will likely by placed at a standoff of 1 m, a tally of the neutron spectrum crossing a 12.7 cm disk at a standoff of 1 m was calculated. Detector response will be considered at a later time.

The first simulation considered vacuum in the volumes occupied by $UF_6$ and steel; this case calculates the unperturbed neutron contributions at the tally location. In sequence, the next simulation considered turning the 30B cylinder volume back to steel, then the source volume into $UF_6$ (but without induced fission). Finally, prompt fission and then delayed fission contributions were added. Figure 4 displays the results of this study. The biggest effect seems to be scattering of high-energy neutrons by $UF_6$, followed by downscattering on steel and induced fission.

Simulations of each of the three enrichments considered were run in the MCNP5 model using a number of starting neutrons corresponding to a very long measurement (10 h) for good statistics. (Note that the number of starting neutrons was different for each simulation, as the (α,n) intensity increases with enrichment due to the larger concentration of $^{234}U$.) The tallied neutron flux through a 12.7 cm-diameter detector face at a standoff of 1 m is plotted in Figure 5.

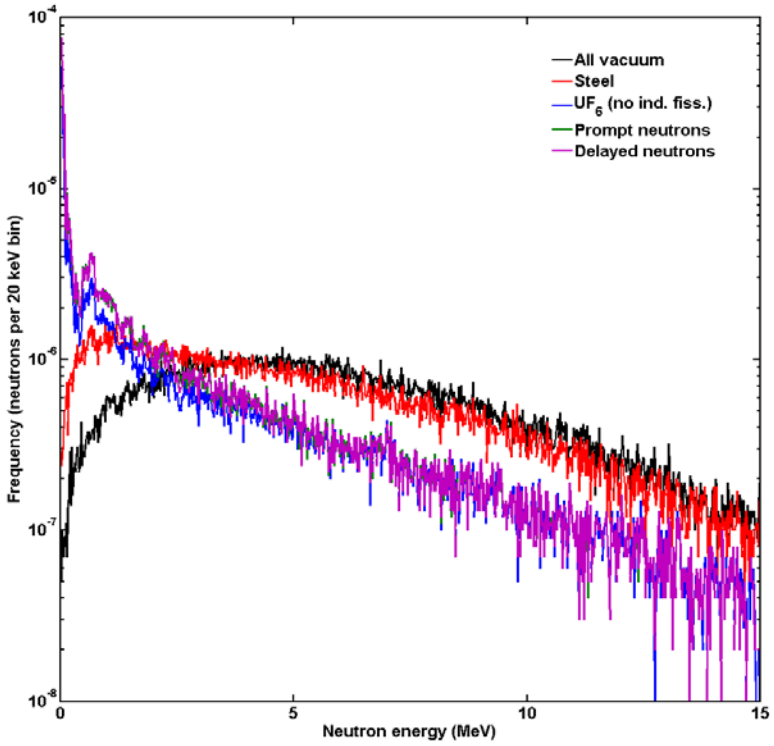


**Figure 4. The effects of scattering, capture, and induced fission on the neutron energy spectrum emitted from the cylinder for a $^{nat}$UF6 source.**

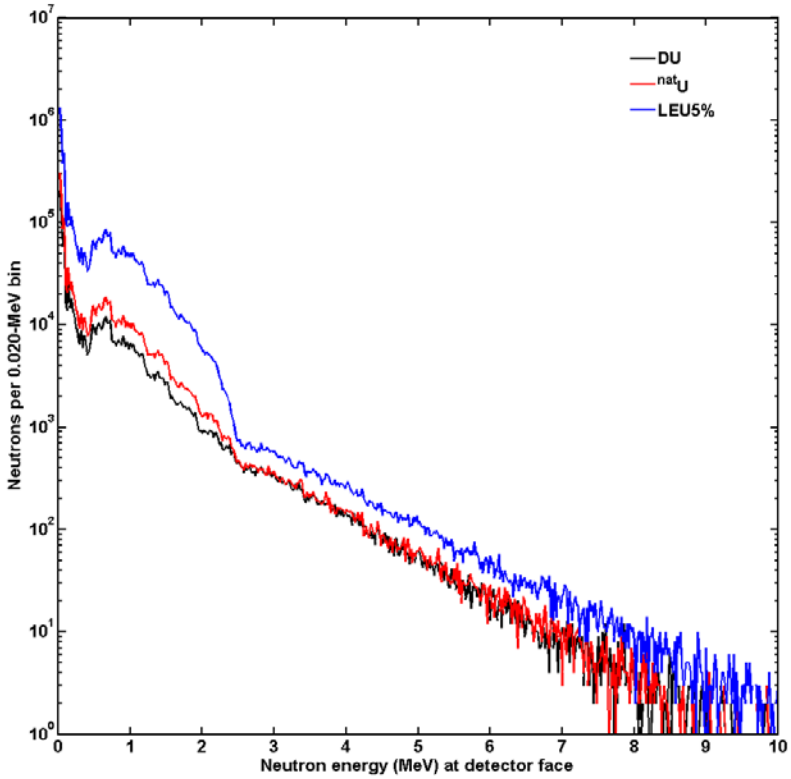


**Figure 5. The calculated neutron flux entering a single 12.7 cm-diameter detector element at a standoff of 1 m.**

A more reasonable measurement interval of 1800 s has also been considered (remembering that this flux is for only a single detector element—the proposed detector has many such elements), and data is used to form Figure 6. This figure presents the total counts above and below 2.54 MeV, which is the location of the knee at the upper end of the (α,n) spectral region, to those from 1 to 2.54 MeV (the detection threshold will likely be near 1 MeV). From the limited data considered so far, it does appear that the bin ratio is a monotonic function of enrichment, thereby lending credibility to the neutron spectrometry enrichment measurement technique.

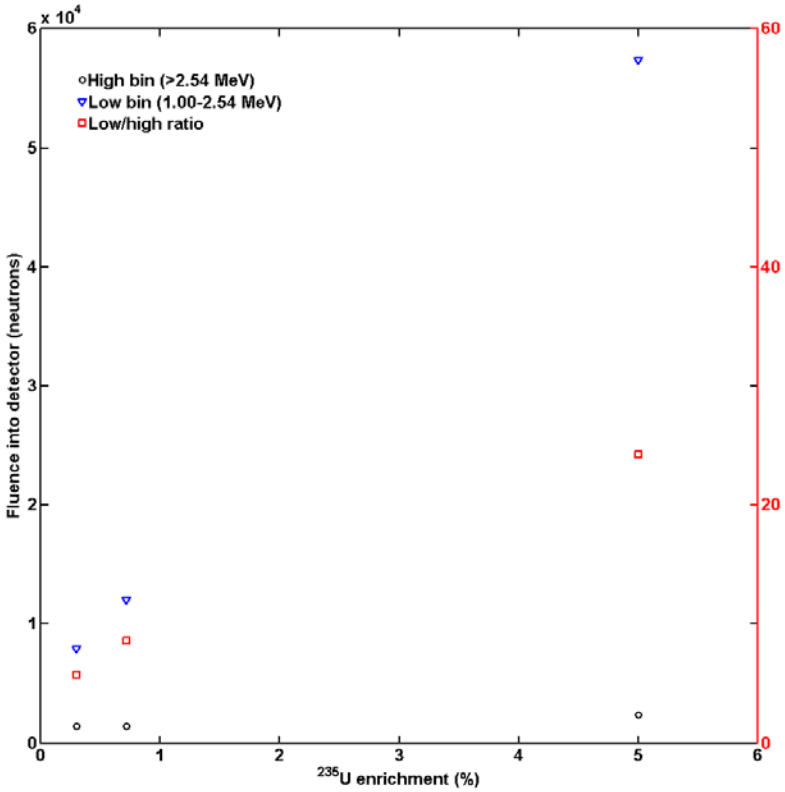


**Figure 6. Left axis: the total number of neutrons entering a single 12.7 cm-diameter detector in the high energy (>2.54 MeV) and low energy (1-2.54 MeV) spectral bins for a 30-minute measurement interval. Right axis: the low/high bin ratio. The spectral ratio is shown to increase monotonically with enrichment for the data considered.**

## DISCUSSION

Neutron spectroscopy has been accomplished at Sandia using the neutron scatter camera [6], which can be used for initial uranium enrichment measurements under this project. The neutron scatter camera is an array of detection cells filled with organic liquid scintillator, which is used because pulse shape discrimination can be implemented with this detection medium to distinguish neutrons from gamma rays that can contaminate the desired signal. In the neutron scatter camera, the total energy for each event is calculated as the sum of two components: the energy deposited in the first cell (calculated from the measured scintillation pulse amplitude) and the energy of the neutron as it leaves that cell, calculated from the time-of-flight between the two triggered cells.

As currently configured the neutron scatter camera is optimized to locate sources in far-field imaging, and some modifications of liquid scintillator cell sizes and spacing may be required for 30B cylinder enrichment measurements. It may very well be that the neutron scatter camera is not the ideal neutron spectrometer for this application, as it currently has a detection threshold for double-scatter events near 1 MeV—as seen in Figure 5, this high threshold will cut out a large portion of events, which may reduce the measurement accuracy or increase the required measurement time. A survey of other neutron spectrometry technologies is planned for the future.

There are, of course, limitations to this measurement concept. The measurement precision is unlikely to ever challenge the performance of the enrichment meter [7], although the point of the neutron spectrometer is to reduce potential systematic uncertainties, which could potentially be large with the enrichment meter in uncontrolled measurement configurations. Also, it is known that the gamma-ray flux emitted from 30B cylinders can be quite large; for the neutron scatter camera, which is sensitive to gamma rays, the gamma-ray flux may create a large system dead time unless preventative measures are taken, like using lead shielding. Finally, the neutron spectrometry technique relies upon a well-characterized relationship between $^{234}U$ and $^{235}U$ isotopic fractions; however, the introduction of reprocessed uranium may complicate the analysis because the isotope ratios will be different after sufficient neutron irradiation in a reactor. It should be stated that these potential concerns are not unique to the proposed neutron spectrometry enrichment measurement: the gamma ray flux is a concern for all detectors except those that are truly gamma-insensitive (such as $^{3}He$ proportional detectors), and the reprocessed fuel will be a potential complication for any neutron-based measurement (including measurements based upon the conversion of neutrons into gamma-rays). There are plans to investigate all of these topics in relation to the neutron spectrometry technique.

Finally, it is worth mentioning some minor benefits of utilizing the neutron scatter camera for the neutron spectrometry enrichment measurement. The neutron scatter camera is capable of neutron imaging, and it is possible to reject backgrounds—either a natural background or from nearby 30B cylinders—by performing neutron imaging during the measurement and rejecting neutrons that originate outside the region occupied by the cylinder being interrogated. Also, neutron imaging can potentially be used to image the $UF_6$ distribution within a 30B cylinder, which could provide a safeguards verification tool allowing inspectors to determine whether the cylinder has internal structure that is unexpected.

## CONCLUSIONS

In summary, this paper has provided the basis for a uranium enrichment measurement using neutron spectrometry. Simulations of 30B cylinder emissions have demonstrated that the source neutron flux emitted from the $UF_6$ material is perturbed during transport through the $UF_6$ and cylinder wall, although the neutron flux reaching the detector still contains an energy dependence related to the $^{235}U$ content of the cylinder. Measurements are planned for the future using Sandia's neutron scatter camera, which has demonstrated neutron spectroscopy in past measurements.

## REFERENCES


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