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SIMPLE, AUTHENTICABLE MEASUREMENT OF FISSILE MATERIALS

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ABSTRACT

Future nuclear weapons treaties may limit the number of warheads a party possesses, which will require verification measurements of those warheads. To allow such measurements, innovative methods are needed that balance confidence in a measurement with safety and information protection. To this end, we are developing methods for measuring fissile materials using a moderated neutron source and superheated droplet (bubble) detectors. The moderated neutron source provides neutrons of energy below the fission cross-section of fissionable materials, such as ^{238}U and ^{240}Pu, so that the resulting fission neutrons are due to interactions with the fissile material in the interrogated item, such as ^{235}U and ^{241}Pu. Droplet detectors are well-suited to this measurement since they are gamma blind and can be designed to have an energy threshold to minimize the detection of the moderated neutron source. Furthermore, measurement of the droplet detector response to the neutron flux can span from qualitative, using human senses to see or hear the bubbles being generated, to quantitative, counting bubbles with image processing and even spectral measurements with multiple neutron thresholds. Here, we present simulation studies to determine the feasibility of the fissile material measurements, including the design of the moderated neutron source to achieve an acceptable neutron spectrum and flux and initial studies of the response of droplet detectors from two suppliers.

INTRODUCTION

Future nuclear weapon treaties are likely to limit the number of warheads, which will require verification measurements of those warheads. Approaches that are safe, intrinsically protect information, and provide a trusted result confirming a physical property fundamental to a nuclear warhead are needed. The protection of warhead design information is a critical aspect of these measurements, although the nature of the information protected varies with the objectives of the treaty. For instance, a warhead-inventory-limiting treaty between two nuclear powers is likely not to allow disclosure of the amount of fissile material in each warhead, but a material accountancy treaty will quantify the fissile mass of the warhead while protecting other aspects of the design, such as geometry. A simple approach that can be trusted by both parties to protect information and provide reliable results is more likely to be adopted in a verification regime than a high-intrusion

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measurement, which involves measurements that require an electronic information barrier for security.

There is no generally accepted system for confirming that a treaty-accountable item is a nuclear warhead. Concepts for monitoring regimes range from less-intrusive approaches, e.g., documentation and dose-rate measurements, to highly-intrusive NDA measurements, e.g. gamma spectroscopy potentially coupled with neutron multiplicity. The less intrusive approaches provide lower confidence to inspectors as they do not provide information that is specific to the intrinsic properties of nuclear warheads. For example, spontaneous neutron emission could be produced by a simple ^{252}Cf source. The highly intrusive approaches require an information barrier to protect information acquired during the measurement that cannot be released. The information barrier then presents both certification concerns for the host, e.g., is it absolutely effective at protecting sensitive information, and authentication concerns for the inspector, e.g., how to have confidence equipment is reporting information correctly.

One of the most developed and tested systems for arms control verification is TRIS, the Trusted Radiation Identification System [1]. TRIS is a NaI-based gamma-ray templating system which requires an initial measurement on a trusted item and compares subsequent measurements to that trusted spectrum to confirm that they are similar. There are several challenges with TRIS. As with all templating systems, one must have confidence that the trusted item is authentic. As a gamma spectroscopic system, an electronic information barrier is required. The presence of high-Z materials and self- attenuation may reduce the critical gamma-ray signatures. Furthermore, the system is most sensitive to material at the outer edge of the fissile components. The handling of the information with TRIS, from generating encryption keys to generating the original template, to handling of that data, is a long series of steps, that while each step individually is not complex, collectively they may be viewed as operationally burdensome.

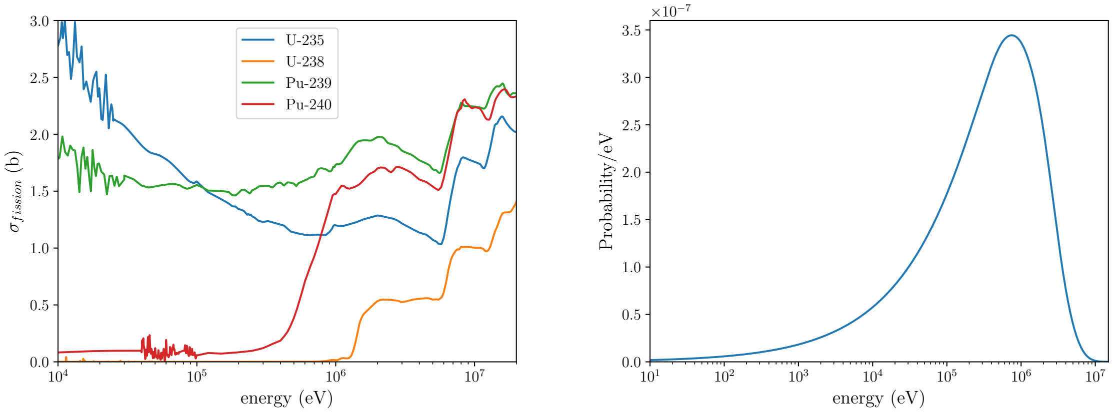

Figure 1. (left) Fission cross-sections for a variety of nuclear materials, including fissile ^{235}U and ^{239}Pu. (right) The fission emission spectrum from ^{235}U due to a 500-keV incident neutron.

This work will study treaty verification of fissile material in a nuclear warhead that protect information and provide a trusted result. Our fundamental concept is to irradiate treaty-accountable items with sub-MeV neutrons and measure the fission driven by this irradiation by observing fission neutrons with more than an MeV of energy. Three different applications will be studied:

1. Attribute measurements – to determine the *presence* of fissile material,

2. Template measurements – to *compare* two objects and confirm consistency, and
3. Accountancy measurements – to *quantify* fissile material mass.

The work presented here focuses on the attribute measurement, while other work presented at the 2021 INMM/ESARDA meeting discusses progress on making template measurements using a film-based zero-knowledge protocol [2].

The attribute measurement approach uses a detector system that can be “read out” with the human senses. This “read out” approach limits the nature of the measurement to a qualitative one, which protects information. The signature for the presence of fissile material is a qualitative increase in the signal when fissile material is present compared to when it is absent. The detector must have an energy threshold to distinguish source neutrons from fission neutrons generated in the fissile material. The energy distribution of the fission neutrons is shown in Figure 1 (right). The detector must also be capable of being “read out” through visual or audible means that are non-quantitative. The background sources for the attribute measurements will be neutrons above 1 MeV that are generated on the same size of depleted uranium (DU) (0.25% ^{235}U). Those background neutrons can come either directly from the neutron source, or through fission neutrons generated in the DU by the small fraction of source neutrons above the ^{238}U fission threshold, see Figure 1 (left). The neutron signal induced in the fissile material must be strong enough to be qualitatively distinguished from these background neutron rates.

This paper describes progress towards making a measurement with a moderated neutron source to differentiate a target with a significant fraction of fissile material from that without. We describe measurements of the response of droplet detectors to a few common neutron spectra, detail modeling efforts of the moderated neutron source to confidently measure fissile materials, and discuss plans for measurements in the upcoming year.

DROPLET DETECTOR RESPONSE BENCHMARK MEASUREMENTS

Droplet detectors are a candidate for the attribute measurement. They are simple, nonelectronic, and can be read-out by eye, specifically by counting bubbles. They have been applied to a variety of detection problems [3-5], including detection of special nuclear materials in arms control applications [6, 7]. A further benefit is that they are insensitive to gamma radiation.

Droplet (so-called “bubble”) detectors contain superheated droplets in a liquid medium, in which the droplets are above their boiling temperature but maintained in liquid phase by external pressure and surface tension. When a sufficiently energetic neutron interacts in the superheated droplet, it creates a small gaseous cavity. When this cavity is large enough the droplet becomes fully vaporized, creating a visible bubble. Examples of droplet detectors that have been exposed to a neutron fluence are shown in Figure 2. The bubbles are clearly visible and can be counted by eye if needed.

For the experiments, droplet detectors have been acquired from two suppliers, Bubble Technology Industries (BTI) and Yale University. From BTI, two varieties of the Bubble Detector Spectrometer detectors were purchased, with a nominal energy threshold at 600 keV and 1 MeV. From Yale, some of these detectors have a higher neutron detection efficiency due to a higher droplet density than the others.

Figure 2. Droplet detectors showing bubbles formed after being exposed to ^{252}Cf.

To better understand the droplet detector response, we exposed the detectors to AmBe and ^{252}Cf neutron sources, where the ^{252}Cf source was configured with and without heavy water moderation. These sources offer three unique spectra for developing and benchmarking a detector response model that describes the number of bubbles generated in a droplet detector. Initial results confirm that an increase of neutron efficiency of about an order of magnitude, from 10^{-4} to 10^{-3} [bubbles/(neutrons/cm^2)], is realizable with the Yale detectors. This improvement in efficiency directly affects the feasibility of the measurement in modeled studies, as described in the following section. Work is continuing to understand the droplet detectors response to these spectra.

MODELING FOR MODERATED SOURCE IMPROVEMENTS

An existing moderated neutron source at the Princeton Plasma Physics Laboratory (PPPL) is a good starting point for the planned attribute measurements. This source, EXCALIBUR ("**Ex**periment for **Calib**ration with **Ur**anium") uses steel moderation to reduce the energy of ~14 MeV neutrons from a deuterium-tritium (DT) neutron generator to sub-MeV energies. A drawing of this neutron source is shown in Figure 3 [8]. Other than the steel moderator and surrounding borated polyethylene shielding, the drawing also shows a nominal location for objects to be inspected.

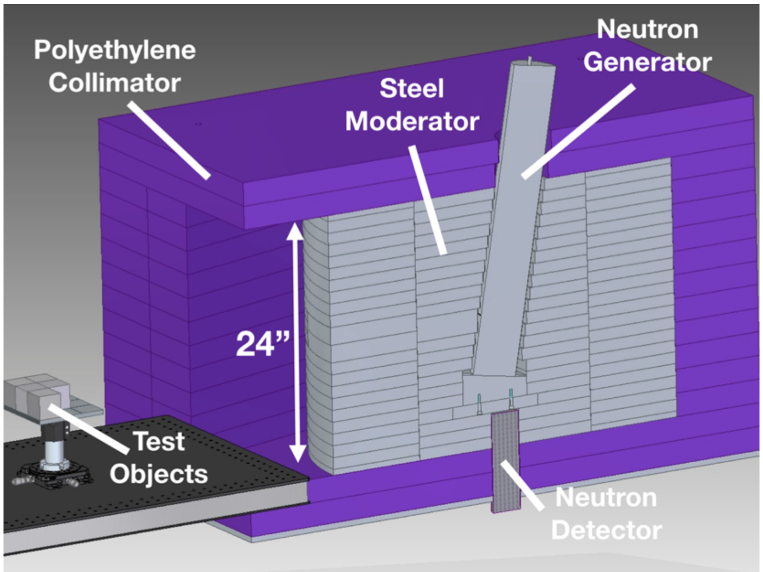

Figure 3. The baseline moderated neutron source located at the Princeton Plasma Physics Laboratory.

For the attribute measurement, the signal of interest is fast neutrons ($\gtrsim$ 1 MeV) from fission generated by sub-MeV neutrons that induce fission in the fissile material of the target. The dominate background contributions are fast neutrons coming from the source to the detector with minimal scatter and fast neutrons that induce fission in the fissionable material of the target. Ideally, the second background term would not exist, but in practice no moderator of 14-MeV neutrons can eliminate high-energy neutrons while providing adequate neutron flux. Since the measurement is qualitative, we consider the number of bubbles that are needed to show a qualitative difference between a fissionable material target (e.g., DU) and a fissile material target (e.g., High-assay low-enriched uranium with ~16% ^{235}U). We assume that a qualitative difference between two bubble detectors can be confidently established if:

1. there are fewer than 100 bubbles present in the detector, since more bubbles cause occlusions, and
2. there is a factor of at least 1.5 between the mean number of bubbles generated in the target material and the background material.

The number of bubbles required in the signal measurement to confidently discriminate the signal measurement from the background measurement depends on the required level of separation between the two measurements. Table 1 summarizes the maximum number of bubbles in the background given a fixed signal count that provides a 90% confidence that the number of bubbles in the signal and background measurements are at least a factor of 1.5 or 2.0 different. These calculations assume that both the signal and background are Poisson-distributed. From this table we see that higher signal-to-background ratios require fewer signal counts and that a higher required separation factor, 1.5 vs 2.0, requires a higher signal-to-background for the same signal count. From this we observe that for confidence in observing a difference of 1.5x, the measurement the separation between the number of signal counts and background counts is closer to 2x.

Table 1. Maximum mean background counts consistent with a given mean signal count in which there is a 90% likelihood that the measured signal to measured background counts is 1.5 and 2.0.

SIGNAL COUNTS	MAX. BACKGROUND COUNTS	
	Factor = 1.5	Factor = 2.0
20	8	5
40	19	13
60	30	22
80	42	31
100	54	39

We are targeting measurements next year that demonstrate the ability to distinguish a 2-in x 2-in x 2-in block of high-assay low-enriched (~16% ^{235}U) uranium (HALEU) from a block of the same dimensions of depleted uranium (DU) with 0.25% ^{235}U. Admittedly, this is more difficult than distinguishing highly-enriched uranium from low-enriched uranium but necessary for ease in executing the experiment.

We simulated in MCNP [9] the baseline moderated neutron system that was optimized by Hepler [8]. For the attribute measurement in comparison to the template measurements, it is particularly important to minimize the neutron flux for energies greater than 1 MeV coming from the source. A simple argument to support this notion is the difference in the level of counts required for the two measurements, O(10) for the attribute measurements and O(1000) for the template measurements; the higher statistics enable observing a smaller difference between signal-and-background for the template measurements and achieving higher discrimination in the face of both significant shielding and the requirement to have a high preload, consistent with the ZKP approach. The reduction of neutrons from the source with energies greater than 1 MeV will reduce both the high-energy flux passing from the source directly to the detector and the fissions produced in the fissionable DU cube. We found that the original source design provided a ratio of the mean signal to background counts of 1.51, for bare HALEU vs. DU. A measurement to achieve 90% confidence that a signal and background measurement are at least a factor of 1.5 apart is extremely long (many, many hours), so that it is not practical to use the existing EXCALIBUR design for the attribute measurements.

We considered several modifications to improve the performance of the neutron source for the attribute measurements. The result of this optimization, making slight modifications to the original design, is shown in Figure 4. First, a 2-in thick beam filter of polyethylene was placed between the source and the target. This filter lowers the energy of the flux incident on both the target and the detectors, and reduces the unattenuated 14-MeV neutron flux, producing more fission in the target and reducing background in the detectors. Second, a steel shadow bar between the source and the detector, which reduces the high-energy neutron flux coming directly from the source, was increased to 30-cm long x 64-cm wide x 21-cm deep. Third, the detectors were moved 5-cm back (away from the target material), which also reduces the background but reduces the efficiency for detecting neutrons generated in the target.

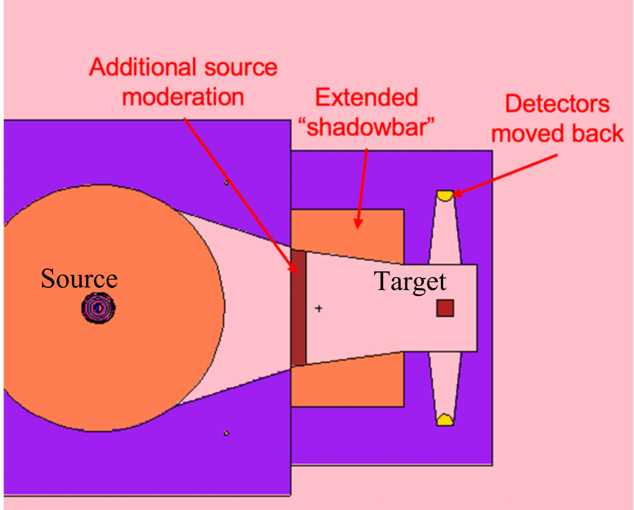

Figure 4. Top-down view of the optimized moderated source design. Neutrons produced in the source on the left are moderated in the nearby steel (orange) and travel to the right towards the target material. Borated polyethylene is shown in purple and polyethylene is shown in dark red.

The updated system, shown in Figure 4, significantly improves the signal from the HALEU compared to background from the DU, but at the cost of lower bubble generation rate. The improvement in performance from the original to the revised design is summarized in Table 2. Though fewer bubbles in a given time are produced with the revised system, a confident measurement can be made quicker due to the larger separation between the HALEU and DU signals. The absolute neutron rates assume a bank of 48 droplet detectors and a DT generator of yield $2 \cdot 10^8$ n/s. The number of bubbles was determined by multiplying the neutron spectrum at the detector to the energy dependent efficiency given in the literature [3], which are approximately 10^{-4} above the energy threshold. With the droplet detectors from Yale, higher efficiencies by an order of magnitude can be realized. However, in this case more bubbles per detector may not be advantageous. It may be easier to count many detectors with 0-2 bubbles than few detectors with many bubbles.

Table 2. A summary comparison of the signal from a target with fissile material (HALEU) and that from a target with much less fissile material (DU).

	BUBBLES/HOUR	
	Original	**Revised**
HALEU	123	46.8
DU	81.9	18.6
HALEU/DU	**1.51**	**2.52**
Time for separation of DU/HALEU	~Hours	30 minutes

CONCLUSIONS AND FUTURE WORK

We will demonstrate the ability to measure the presence of fissile material with simple, nonelectronic neutron detectors and a moderated neutron source. We have acquired droplet detectors and are currently making measurements with a variety of neutron spectra. From these results, we will build a detector response model, which determines the number of bubbles in the detector for a given neutron spectral flux.

In preparation for the demonstration measurements next year, we are completing a modeling study to determine simple modifications that can be made to an existing moderated neutron source to support the measurement. Our objective is to demonstrate measurements on HALEU and DU with a visible distinction in the detector response for the two measurements. We determined that confident measurements can be made with limited modifications to the existing system. This indicates that demonstration measurements can be made in the upcoming year. The demonstration measurements are planned for 2022.

Success in the measurements will provide a pathway to simple, authenticable fissile material measurements. Importantly, the interrogation approach is built considering trust of the host and confidence of the inspector, providing another tool in the toolbox in negotiations over future arms control treaties.

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