Zero-Knowledge Differential Isotopic Comparison of Special Nuclear Materials

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ABSTRACT. Zero-knowledge proofs are mathematical cryptographic methods to demonstrate the validity of a statement while providing no further information related to the statement itself. The possibility of using such proofs to process sensitive physical data without ever acquiring sensitive information has gained attention for its potential application to nuclear warhead or subassembly inspection. Recently, we have provided experimental evidence that measuring sensitive data is not required to perform comparisons of physical properties, in particular opacity to 14 MeV neutrons. Here, we report on the development of a non-electronic active neutron interrogation technique using superheated emulsion detectors to discriminate special nuclear materials of identical geometry but different isotopic composition, without learning what these properties are.

Introduction

Techniques to confirm the authenticity of nuclear weapons without sharing sensitive information could facilitate the negotiation and verification of future nuclear arms reduction and disarmament treaties. Following the work of Glaser, Barak and Goldston,1 we have demonstrated a physical zero-knowledge system based on a non-electronic neutron differential radiography technique to perform reproducible object comparison without ever acquiring data about the objects being compared, if they are identical.2 Experimental results, with 14-MeV neutrons and non-fissile test objects, established a physical basis for the implementation of such a system as part of an interactive proof protocol to confirm that two nuclear warheads are identical without learning sensitive geometry and material composition information.3 Current efforts focus on applying our technique to the discrimination of plutonium and uranium objects of different isotopic composition and configuration, without learning what these properties are.

In this paper, we report on the current status of these efforts. First, we review and discuss practical approaches to actively interrogate and discriminate special nuclear materials using different neutron energy sources and non-electronic super-heated droplet
(bubble) detectors. Then, to validate theoretical and computational results, we propose experiments involving the active interrogation of kilograms quantities of highly enriched uranium, as well as an upgrade of our experimental apparatus at the Princeton Plasma Physics Laboratory (PPPL) to measure 14-MeV induced neutron emissions at large angles from natural and low enriched uranium objects.

**Active Neutron Isotopic Discrimination of Fissile Materials**

Our approach to warhead comparison consists in exposing objects to a neutron beam and recording their neutron transmission and emission patterns on a set of non-electronic super-heated droplet (bubble) detectors (Figure 1). By comparing the neutron counts in each detector, we seek to distinguish objects of different geometry and composition.

![Figure 1](image)

*Figure 1.* Current set-up of our physical zero-knowledge comparison system located at the Princeton Plasma Physics Laboratory. A collimated 14-MeV neutron beam illuminate 2 inch cubes (not on the picture) of different materials and configuration arranged on a 3-by-3 grid. Bubble detectors record both neutron transmission and emission (at 90°) signals.

By preloading our detectors with complement radiographs of the inspection patterns before irradiation such that the sum of these signals is equal to the expected signal, $N_{max}$, our system can show that two objects are identical without learning what they are (zero-knowledge property). The expected signal $N_{max}$ can correspond, for example, to the signal recorded with non-preloaded detectors when no object is present between the neutron source and the detectors.
To construct a proof of physical equality between two objects from a proof of neutron radiograph equality, we first assume that if the objects are not significantly different, then the neutron counts recorded by the detectors at the end of an inspection must be statistically close for all orientation and neutron source energy pairs \((\theta_i, E_j)\), \(i = 1...n\) and \(j = 1...m\), with probability at least \((1-\alpha)\). Furthermore, we make the physical assumption that there exists \(\beta < 1 - \alpha\) such that for any object \(X\) and any object \(Y\), if \(Y\) significantly differs from \(X\), then there exists an orientation-energy pair \((\theta_i, E_j)\) such that testing with these parameters will lead to a significantly different neutron count with probability at most \(\beta < 1\).

We have shown computationally and experimentally that 14-MeV neutron transmission radiography with non-electronic super-heated emulsion detectors is sensitive to geometry and elemental differences of test objects.\(^4\) We have also shown that matching an object with a valid complement radiograph leads to the expected zero-knowledge result with a well-understood level of noise. Fast neutrons of 14-MeV energy, however, are not always sufficient for discriminating between fissile and fissionable material in driven emission measurement. In particular, the fission cross-sections, which dominate the total cross-section at energies above 10 MeV, can be very close for isotopes of uranium and plutonium (Figure 2).

![Figure 2. Fission cross sections of uranium-235, uranium-238, plutonium-239 and plutonium-240 with average energy of relevant neutron sources.](image-url)

To address this issue, we have proposed to use a \(\sim 250\)-keV neutron source based on the \(^7\text{Li}(p,n)^7\text{Be}\) reaction, which has a threshold at 1.88 MeV and a resonance near 2.25 MeV.\(^5\) Yan and Glaser studied the viability of this “two-color” neutron detection approach computationally.\(^6\) Using detectors with different neutron energy thresholds,
they showed that a ∼250-keV p-Li neutron source can detect changes in isotopic composition of uranium through fission emission measurements with 500-keV threshold detectors, which are insensitive to the source neutrons.

In addition, we have proposed the use of large-angle detectors shielded from the collimated beam and sensitive to all neutron producing reactions in the objects being interrogated that can be isotope specific. For uranium objects, it is still possible, using this approach and 14-MeV neutrons, to distinguish significant changes in isotopic composition of a binary mixture of uranium-235 and uranium-238 in kilogram quantities. This is not the case for plutonium where the fission cross sections at 14 MeV are too close. Plutonium isotopes present, however, large differences in spontaneous fission rates that could also be measured.\textsuperscript{7}

To validate experimentally key aspects of these two approaches, we designed a series of new experiments. The first experiments consist in driving fission in the Rocky Flats Shells,\textsuperscript{8} an assembly of nested highly enriched uranium metal spherical shells, with ∼500-keV neutrons from the $^{241}$AmLi($\alpha$,n)$^{10}$B reaction as well as 2.5-Mev DD neutrons using in both cases detectors with thresholds higher than the respective source energy. The other experiments are based on an upgraded version of our existing 14-MeV apparatus to measure large angle neutron emissions from cubes of various materials including low enriched and depleted uranium metal.

**Experiments with the Rocky Flats Shells**

The Rocky Flats Shells (RFS) experiments seek to demonstrate two key points of our approach to zero-knowledge objects comparison: first, we want to show that it is possible to measure fission neutrons emitted from uranium objects that have energies higher than the interrogating neutron source. Second, we want to show that such measurements are sensitive to changes in assembly configuration and isotopic composition, both affecting the neutron reproduction factor $k$, particularly when the source energy is lower than the fission threshold in uranium-238.

The RFS used in these experiments consist of 12 pairs of nested hemispherical shells of uranium metal (1.02 wt% $^{234}$U, 93.16 wt% $^{235}$U, 0.47 wt% $^{236}$U, and 5.35 wt% $^{238}$U) corresponding, when fully assembled, to a solid sphere with a radius of 5.67 cm and a mass of 13.7 kg. The measurements are designed to be independent of the object orientation using the spherical symmetry of the RFS.

Figure 3 shows a representation of the AmLi version of the experimental setup with 10 specially made superheated droplet detectors placed 30 cm away from the uranium sphere. The detectors comprise glass vials (test tubes) filled with an emulsion of fluorocarbon superheated droplets homogeneously dispersed in an aqueous gel matrix.\textsuperscript{9}
In the AmLi experiments, we use octafluorocyclobutane (C-318, $C_4F_8$) to obtain a threshold between 1 and 2 MeV. The DD experiments require a higher threshold to be insensitive to 2.5-MeV neutrons, in this case we use perfluorobutane (R610, $C_4F_{10}$). In both types of detectors, there are $\sim 4000$ droplets per cm$^3$ of 100 µm and 120–130 µm diameter, respectively.

Figure 3. Active neutron interrogation of the Rocky Flats Shells with 4 Am-Li sources and fission signature recorded with superheated emulsion detectors (CAD model).

In the AmLi interrogation experiments, four AmLi neutron sources, each emitting $2.5 \times 10^5$ neutrons per second, are placed symmetrically around the RFS (Figure 3). We modeled the experiment using the Monte Carlo neutron transport code MCNP6. The energy distribution of the source neutrons is modeled from the ideal AmLi spectrum. Simulations consisted in modeling the total bubble count from all 10 detectors during a series of 4-hour measurements. In one scenario, we modeled the removal of uranium from the center of the sphere, shell by shell, keeping the outside radius fixed. In a second scenario, we modeled a solid sphere of uranium metal (binary mixture of uranium-238 and uranium-235), the size of the fully assembled RFS (Shells 1 to 24), and varied the weight fraction of uranium-235. The absolute efficiency of the detectors is of the order of $4 \times 10^{-4}$ bubbles per crossing neutron. Results for both scenarios are presented in Figure 4.

For the purpose of illustrating these results, and because we are interested in isotopic
and configuration discrimination, we make the assumption that the outer radius of the shells is known. This allows us to pick the expected total number of bubbles $N_{\text{max}}$ at the end of the inspection of a valid object. We take $N_{\text{max}} = N_0 + 6 \sqrt{N_0} = 472 + 6 \sqrt{472} \approx 600$ with $N_0$ the expected bubble count for a full solid sphere of pure uranium-235. We then calculate the statistic $T_i = (N_i - N_{\text{max}})^2/N_{\text{max}}$ where $N_i = N_{\text{pre}} + N_{\text{insp}}^i$ is the result of an inspection on configuration $i$, the sum of a valid preload $N_{\text{pre}}$ (the complement for the full RFS) and the induced fission signature $N_{\text{insp}}^i$. Next, we compare $T$ to a $\chi^2$-distribution with one degree of freedom to compute p-values. Statistics are shown in Table 1. The results show that the method can detect a removal of 944 g of HEU from the center of the sphere (corresponding to Shells 1 to 4) at the $\alpha = 0.05$ level.

The results from both scenarios also illustrate an important point: this measurement configuration detects changes in the neutron reproduction factor $k$. It cannot, however, tell alone what is affecting such changes, i.e. mass removal at the center or modification of isotopic composition of the full sphere.

We plan to confirm some of these computational results with actual experiments on the Rocky Flats Shells in configurations 1–24 and 13–24. The DD interrogation experiments will have similar geometric configuration for both the RFS and detectors but only one DD neutron generator will be used.

**Figure 4.** MCNP simulations of Am-Li set-up. (a) Total bubble count as a function of central cavity radius of the Rocky Flats Shells. Each point corresponds to a different configuration of the shells. The two hexagonal points indicate planned experiments (Shells 1 to 24 and Shells 13 to 24). (b) Total bubble count for a solid sphere of uranium metal with similar outer radius but varying weight fraction of uranium-235. In both figures, band represent expected 1-σ poisson noise.
Table 1. Statistics for the detection of inner shells removal assuming $N_{\text{max}} = 600$. p-val corresponds to the probability of detection. Assuming a false positive rate $\alpha = 0.05$, changes in configuration can be identified as soon as the first 4 half-shells are removed, corresponding to a mass of 944 g.

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<td>2.17</td>
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Measurements of Neutron Emissions at Large Angles

The purpose of large-angle (or side) detectors is to measure neutron emissions from the objects being actively interrogated. Such measurements are complementary to transmission measurements. In particular, they can help discriminate objects of similar transmission patterns. The opposite is also true: two objects can have similar emission patterns, but different transmission radiographs.

We have previously developed an apparatus to perform 14-MeV neutron radiography at PPPL (Figure 1). Here we propose to upgrade this apparatus to perform both transmission and emission measurements with an adequate level of statistics to distinguish natural and low-enriched uranium objects.

The upgrade consists in three modifications: First, we replace the existing neutron generator with a more compact model (Thermofisher P-385) increasing the neutron yield to $\sim 5 \times 10^8$ neutrons per second, which corresponds to a five-fold increase compared to our previously used generator; second, we modify the neutron shield and collimator adding a substantial amount of steel in and around the imaging plane to reduce the fast neutron flux reaching the side detectors (Figure 5); and third, we add two steel shadow bars on each side of the collimated fan beam to further shield the side detectors.
The last two modifications result in a reduction by a factor of $\sim 100$ between the neutron fluence above 1 MeV recorded by the emission detectors compared to the transmission detectors when no object is present between the source and the detectors (Figure 5).

The objects studied with this apparatus consist of 2-inch cubes arranged in different patterns on a 3-by-3 grid. Table 2 shows the results of MCNP simulations of the upgraded apparatus including all major pieces of equipment and the room in which the experiment is located. In each simulation, nine cubes of the same material are placed 90 cm from the 14-MeV neutron source and exposed for $\sim 50$ seconds. Transmission and emission signals are recorded on super-heated droplet detectors placed 20 cm away from the objects. Results for each materials correspond to the total bubble counts for 5 transmission and $2 \times 10$ emission detectors respectively. They show for example that, while the natural uranium (0.7 wt% $^{235}$U) and low-enriched uranium (19.75 wt% $^{235}$U) objects cannot be distinguished in transmission, the system should be able to discriminate them based on their neutron emission signals above 1 MeV.
Table 2. MCNP simulations of transmission and emission measurements with the proposed upgrade of the PPPL apparatus. Objects of similar geometry (nine 2”-cubes) but different material compositions are irradiated with a collimated DT neutron beam for 4 minutes. Transmission and emission signals are recorded on 5 and 2×10 (10 at each side of the beam) bubble detectors respectively. Emission results are given for two different thresholds.

<table>
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<tr>
<th>Material</th>
<th>Void</th>
<th>NU</th>
<th>LEU</th>
<th>W</th>
<th>Steel</th>
<th>Al</th>
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Conclusion

We have demonstrated a physical zero-knowledge system based on a non-electronic neutron differential radiography technique to perform reproducible object comparison without an inspector ever acquiring sensitive data when inspected objects are identical to a reference object.

Such a technique requires the active neutron interrogation of items in various orientation and at multiple neutron energies. In this paper, we have devised and modeled a set of new experiments focusing on the discrimination of special nuclear materials of different configurations and isotopic compositions. These experiments consist in: 1) the interrogation of highly enriched uranium nested shells and the measurement of their induced neutron emissions with detectors insensitive to the interrogating source neutrons. 2) the interrogation with 14-MeV neutron of natural and low enriched uranium cubes and their discrimination using large angle shielded emission detectors.

The realization of these experiments with uranium objects should demonstrate key aspects of our approach and represent an important step towards the design of a practical zero-knowledge warhead verification system for field inspection and exercises.

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Endnotes


3A detailed description of the protocol is available in S. Philippe et al. (2016), *op. cit.*

4S. Philippe et al. (2016), *op. cit.*


