Time Correlation Measurements of Heavily Shielded Uranium Metal*

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ABSTRACT

Oak Ridge National Laboratory is developing a method to estimate the enrichment of uranium metal when heavily shielded by high-*Z* materials. The method uses fast neutron tomography to estimate the geometry and materials inside the shielding. With the geometry and materials information, the components suspected of being enriched uranium metal are modeled with different enrichments in Monte Carlo simulations. For each modeled enrichment, a simulation predicts the time correlations expected from large fast plastic scintillation detectors following interrogation with a deuterium-tritium neutron generator. The simulated time correlations that best match the measured time correlations are used to determine the actual enrichment. To test the method, time correlation measurements are made on two annular castings that have a 6 in. height, 3.5 in. inner diameter, and 0.75 in. thickness. One casting is 93% enriched, and the other is depleted uranium. Each casting is surrounded by up to three layers of depleted uranium shielding, with each layer being approximately 0.5 in. thick. This paper presents the results of the measurements and compares the results to the simulations.

INTRODUCTION

Many safeguards and security applications require knowledge and frequent verification of the enrichment of uranium metal items. Under normal circumstances, the items are removed from their containers, and their enrichment is determined using low-energy photons. In some situations, it may be undesirable to remove the items from their containers and/or other shielding. In reference [1], the authors discuss the shortfalls of current methods to determine enrichment when the uranium metal item is surrounded by high-Z materials such that there are no detectable γ-ray emissions. Most methods rely on low-energy photons that are easily shielded by high-Z materials. The authors also present a new method that should work in the presence of shielding materials [1, 2].

The new method involves three primary steps. First, the item and shielding are interrogated with 14 MeV neutrons from a deuterium-tritium (DT) neutron generator (described

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in the next section of this paper). The neutrons from the generator are time and directionally tagged via the associated particle technique [3]. During the interrogation, an array of small fast plastic scintillation detectors at the same height as the DT generator measures the arrival time of particles relative to the time that the 14 MeV interrogating neutron was created. In addition, an array of eight large fast plastic scintillation detectors located around the item also measures the arrival time of neutrons and γ-rays relative to the 14 MeV neutron creation time. After the measurement, the second step involves creating a tomographic image using the ratio of the response of the small detectors with the item present to the response with no item present. Creation of the tomographic image is discussed elsewhere in the literature [4] and will not be repeated here. The tomographic image shows the neutron attenuation within the item and the shielding, and the arrangement of the item and its shielding as well as the materials present are estimated from the image. Next, the geometry and materials estimates from the tomographic image are input into simulations using MCNP-PoliMi [5] models in order to predict the time response of the large fast plastic detectors. Those regions suspected of being enriched uranium are modeled with several enrichments. Finally, the model whose simulated time response best matches the measured time response of the large detectors yields the estimated enrichment of the item.

Many parts of the new method have already been demonstrated or explored with simulations to determine if it will work. Grogan et al. demonstrate estimating the geometry and materials from a tomographic image in reference [6]. Since the neutron attenuation coefficients for ^{235}U and ^{238}U are approximately the same, depleted uranium (DU) and highly enriched uranium (HEU) appear the same in a tomographic image. However, the authors have previously presented sensitivity studies with simulations that show that the time signatures for an item and its shielding are unique for differing enrichments [1]. Therefore, if the simulations can accurately predict the time signatures, the method should work. In fact, in a blind test using only simulated data, Swift et al. were able to predict the enrichment of an item when shielded by depleted uranium to within 5 weight percent [7]. To test the ability of simulations to accurately predict the time signatures, measurements of uranium metal with differing enrichments and surrounded with up to ~ 1.5 in. of depleted uranium (DU) were performed at the Y-12 Nuclear Detection and Sensor Testing Center [8] in February 2012. This paper presents the measured time correlations and compares them to initial simulations.

EXPERIMENTAL SETUP

The two uranium metal items used in the experiment are standard unclassified 161 storage castings used at the Y-12 National Security Complex. The annular castings have a ~6 in. height, 3.50 in. inner diameter, and 0.750 in. thickness. One casting is enriched to 93.186 weight percent ^{235}U, and the other is DU. The castings have a mass of ~18 kg and are canned inside 0.025 in. thick stainless steel to prevent contamination.

Each casting is measured bare and with up to three layers of DU annular shields. The shields are shown in Figure 1, and their dimensions are shown in Table 1. The shields were constructed to allow all three to be used in the same measurement and such that each one weighs approximately 40 lb or less. Like the uranium castings, each shield is canned in 0.025 thick stainless steel to prevent contamination.

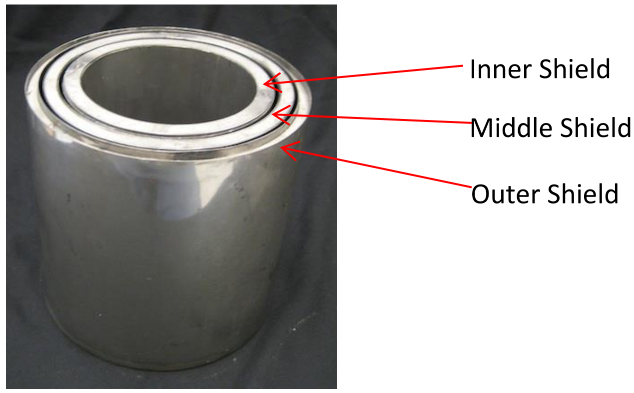

Figure 1. DU annular shields.

Table 1. Dimensions for DU annular shields (not including canning).

Shield	Inside diameter (in.)	Outside diameter (in.)	Height (in.)
Inner	5.300	6.234	6.999
Middle	6.641	7.394	7.011
Outer	7.789	8.394	7.006

An API-120 DT generator manufactured by Thermo Fisher Scientific is used to interrogate the castings and shields [9]. At maximum power, the generator has an output of 3×10^7 neutrons per second. The generator includes an embedded alpha detector, which ORNL helped develop [10]. A pixelated light guide is attached to the alpha detector followed by a Hamamatsu H9500 photomultiplier tube which provides an array of 16×16 pixels. The system uses one row of 16 3.04 mm square pixels. At maximum power, each pixel records ~5000 α particles per second. Since the α particle produced in the DT reaction travels ~180° from the 14 MeV neutron, the detection of an α in one of the pixels gives both the creation time and direction of the 14 MeV neutron.

An array of 32 small fast plastic detectors is located along an arc that is 108.5 cm from the DT generator, as shown in Figure 2. These detectors are used primarily for imaging by detection of neutrons that have passed through the casting without interactions. The dimensions of each detector are 1 in. by 1 in. by 4 in. Pulses from these small imaging detectors are routed to constant fraction discriminators, and the threshold is set such that pulses having less energy than that from the deposition of 1 MeV from a neutron are discarded.

The eight large fast plastic detectors in Figure 2 are used primarily to detect neutrons from induced fission in the uranium casting. The dimensions of each detector are 27 by 27 by

10 cm. To prevent these detectors from interfering with the imaging detectors, the large detectors are placed above and below the line-of-sight from the DT generator to imaging detectors. The bottom four detectors are 28.5 cm above the floor, and the top four detectors are 85.5 cm above the floor. The inner radius for the detector stand is 19 in. To reduce contributions from γ-rays, the front of each detector has a ¼ in. thick lead sheet on it. Similarly, blocks of polyethylene are placed between the detectors to reduce the possibility of a neutron being detected in one detector and scattering into an adjacent detector. In these fission radiation detectors, the threshold is reduced to about 0.5 MeV to allow counting more neutrons.

In the measurements, the casting is centered between the imaging detectors at a distance of 27.8 cm from the DT generator. The mid-height of the casting is 70.5 cm above the floor, which is the height evenly between the top and bottom row fission radiation detectors. The fission radiation detectors are on an arc centered on the casting. The measurements begin with the bare casting and progress with the addition of each shield, beginning with the inner shield. Each configuration is measured for about 35 minutes. Before and after the measurements, the fission radiation detectors were checked with a ^{252}Cf spontaneous fission source to verify that the detectors responded the same throughout the measurements.

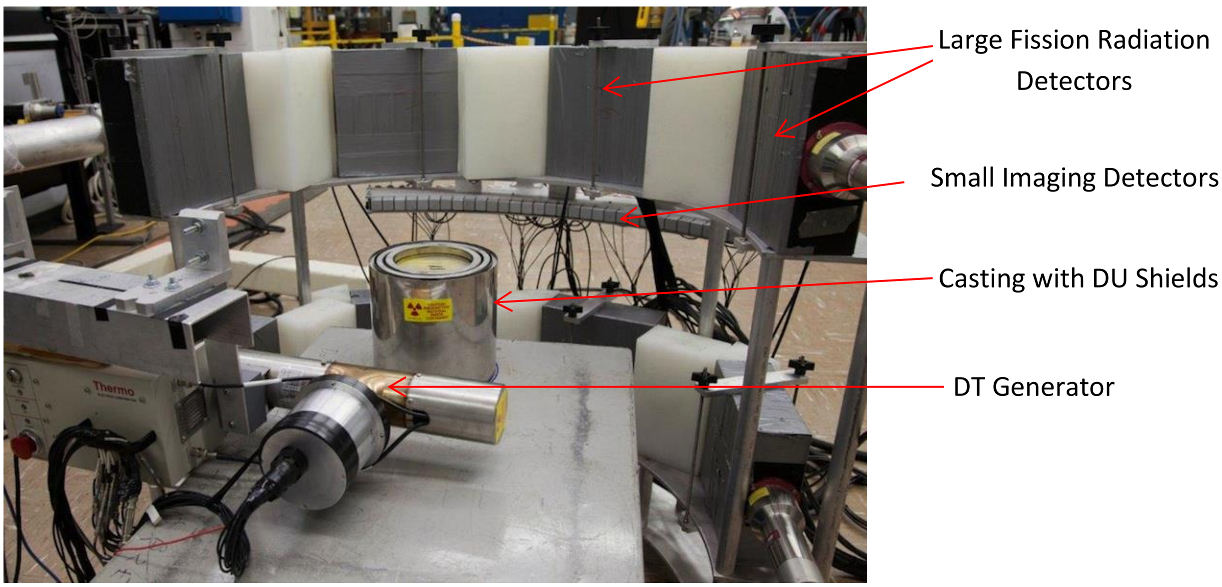

Figure 2. Arrangement of the detector system.

CALCULATIONAL MODEL

The calculational model is illustrated in Figure 3 and is very similar to the experimental setup shown in Figure 1. The model does not include the support stand for the castings and shields, the canning, the support structure for the imaging detectors, nor the internals and walls of the DT generator. However, it does include the walls and floor of the room as well as other structures. Each pixel of the DT generator is simulated with 10 million particles in separate calculations. The probabilistic distribution of neutron intensity and direction for each pixel is modeled with a Gaussian distribution that is estimated from the response of neutron flux seen by the imaging detectors with no object present.

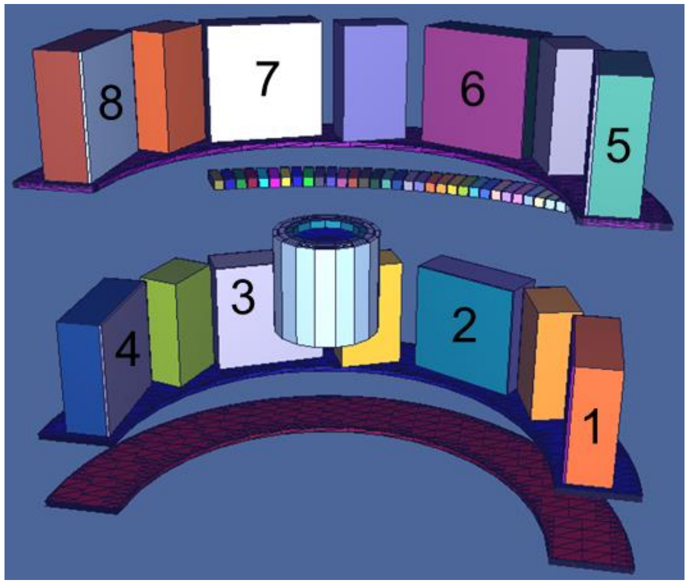

Figure 3. Simulation model.

RESULTS AND ANALYSIS

A comparison of the ^{252}Cf time-of-flight spectrum measured by the fission radiation detectors to the spectrum from the simulations is shown in Figure 4. The color purple in the plot indicates the sum from all the detectors, whereas the color black only shows the composite from the detectors on the top row. Overall, the simulations agree well with the measurements, but there is some disagreement beyond 70 ns where radiation scattered from the room is detected. The simulated spectrum from all the detectors includes slightly more high energy neutrons than the measurement. Since (1) the simulated spectrum from the bottom row detectors is harder than the measurement, (2) the bottom row detectors are more influenced by the floor, and (3) the cart supporting the castings is between the castings and bottom row detectors so that it interferes with the bottom row's measurements, the contribution from the bottom row detectors will be discarded in the following analysis.

The comparison between the measurements and simulations of the castings without any shields is shown in Figure 5. The measurements and simulations of the HEU casting are shown in red, and those of the DU casting are shown in blue. The vertical axis indicates the counts recorded by the detectors on the top row of the stand divided by the total number of α particles recorded by all the pixels. The horizontal axis shows the time after emission of the DT generator neutron. At ~8 ns, γ-rays created from inelastic scattering and induced fission of the uranium castings arrive at the detectors. The peak at ~18 ns is due to 14 MeV neutrons arriving at the detectors. Finally, particles detected after 25 ns are primarily fission neutrons. For the bare casting, the simulations agree well with measurements in the time range of ~35 ns to 60 ns. After

60 ns, the fission neutron tail drops below the actual measurements. At 80 ns, the simulation for the HEU casting is close the measurements for the DU casting.

A comparison of all the shield combinations is shown in Figure 6. The color scheme and symbols in Figure 6 is the same as that for Figure 5. The top of each subplot in Figure 6 shows the shields that are present; that is, “I” is for the inner shield, “M” is for the middle shield, and “O” is for the outer shield. As more shielding is added to the configuration, the simulations begin overpredicting the fission neutrons. For the case where all shields are present, the peak in the fission neutron region for the simulation with the DU casting is close to the measured peak for the HEU casting. In fact, the simulation peak for the DU casting overpredicts the measured peak for the DU casting by 31%.

While Figure 6 shows the comparison considering top row detectors and all pixels of the DT generator, the simulations for some individual combinations of a detector and a pixel have better agreement with the measurements. For example, a comparison of fission radiation detector 5, as labeled in Figure 3, with pixel 11 is shown in Figure 7. While not perfect, the simulations agree with the measurements much better than those shown in Figure 6.

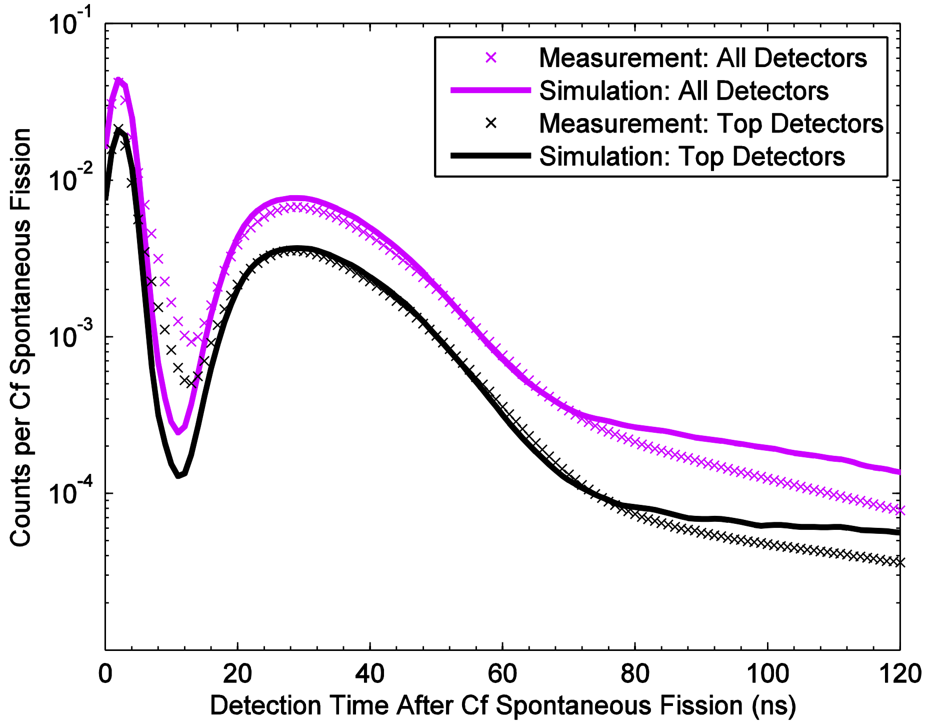

Figure 4. Comparison of the ^{252}Cf measurements and simulations.

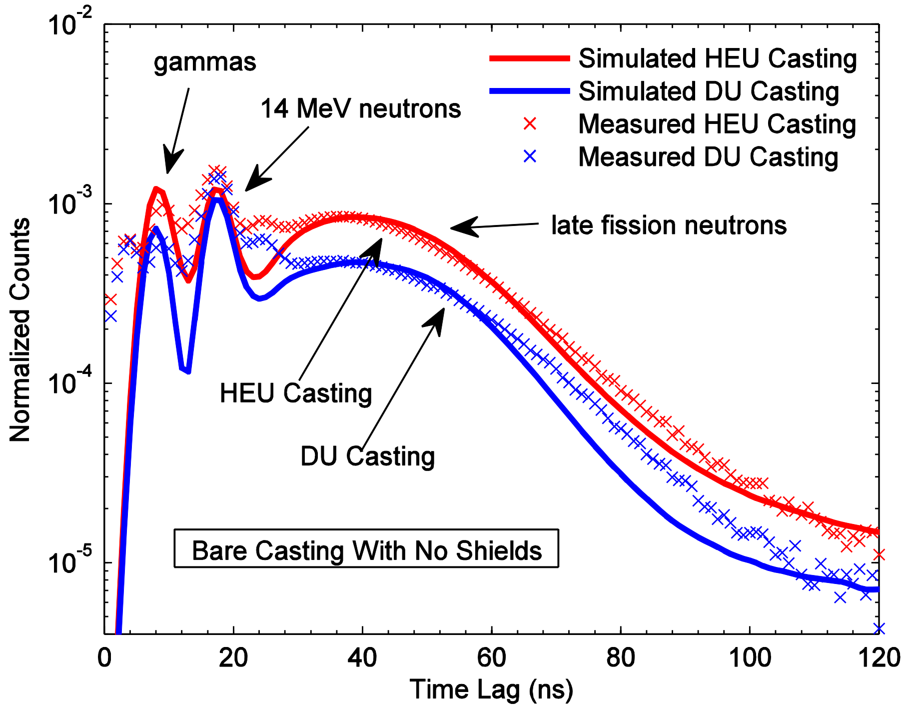

Figure 5. Comparison of the measurements and simulations for the bare castings.

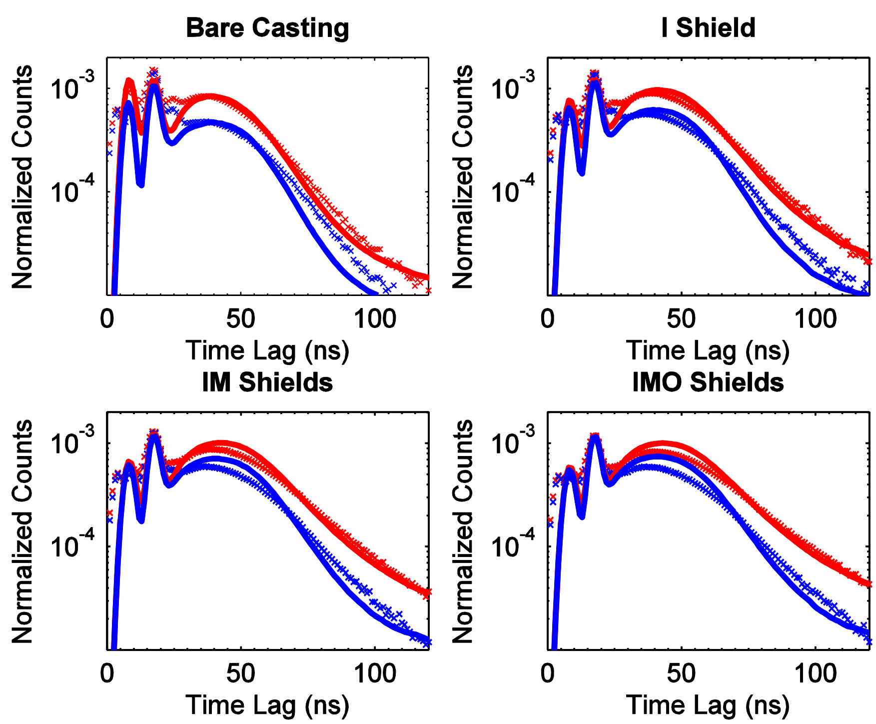

Figure 6. Comparison of simulations and measurements for all the shield combinations using the top row detectors and all pixels of the α detector. The solid lines are simulations, and the x's are measurements.

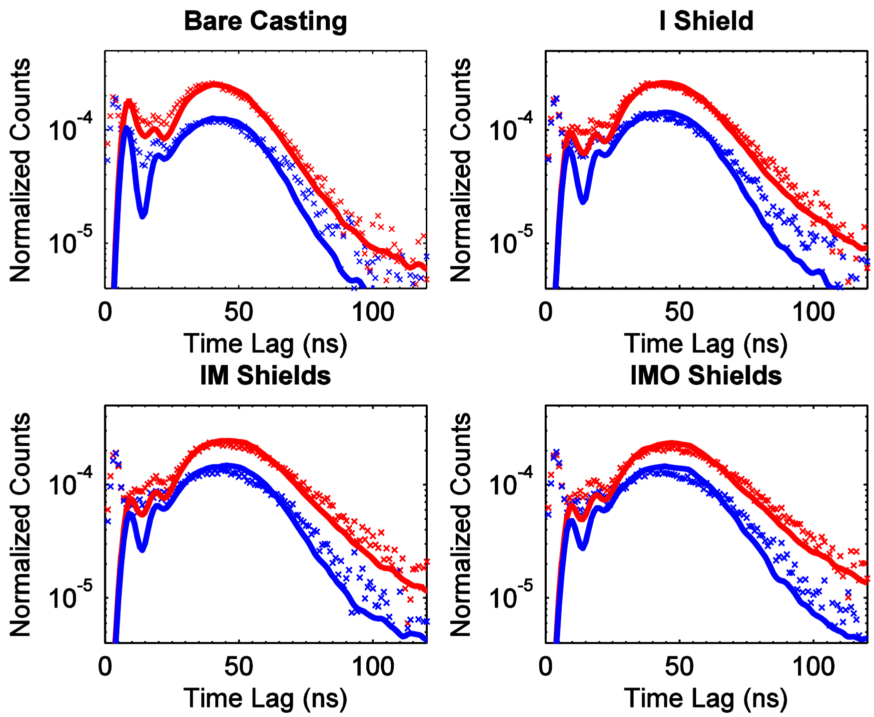

Figure 7. Comparison of simulations and measurements for all the shield combinations using detector #5 and pixel #11 of the α detector. The solid lines are simulations, and the x's are measurements.

CONCLUSIONS AND FUTURE WORK

Comparing the initial simulations to the measurements shows that developing an algorithm to extract the enrichment of uranium metal will be difficult if absolute agreement between the simulations and measurements is necessary. ORNL is currently trying to understand the discrepancy between simulated and measured time correlation observables. First, the current model of each generator pixel's neutron emission beam is a circular cone. In reality, the neutron beam is an elliptical cone, extended in the vertical direction. Efforts are under way to more accurately characterize and model the neutron beam. Secondly, MCNPX-PoliMi, a newer version of the code used for the simulations, was released through the Radiation Safety Information Computational Center in April 2012 [11]. Unlike the older version, the energy of each fission neutron is dependent upon the multiplicity, which should improve the model's agreement with the energy-dependent cross section and time-of-flight effects observed in the measurement. The new version is also able to use Evaluated Nuclear Data Format 7 cross-section libraries. Once the approximations in the neutron beam profile and fission are removed, as well as any additional refinements of the simulation model, the accuracy of this method of enrichment estimation will be evaluated.

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