ENRICHMENT DETERMINATION OF URANIUM METAL IN SHIELDED CONFIGURATIONS WITHOUT CALIBRATION STANDARDS

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To the Graduate Council:

I am submitting herewith a dissertation written by Jason Michael Crye entitled "ENRICHMENT DETERMINATION OF URANIUM METAL IN SHIELDED CONFIGURATIONS WITHOUT CALIBRATION STANDARDS." I have examined the final electronic copy of this dissertation for form and content and recommend that it be accepted in partial fulfillment of the requirements for the degree of Doctor of Philosophy, with a major in Nuclear Engineering.

Howard L. Hall, Major Professor

We have read this dissertation and recommend its acceptance:

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Vice Provost and Dean of the Graduate School

(Original signatures are on file with official student records.)
ENRICHMENT DETERMINATION OF URANIUM METAL IN SHIELDED CONFIGURATIONS WITHOUT CALIBRATION STANDARDS

A Dissertation
Presented for the
Doctor of Philosophy
Degree
The University of Tennessee, Knoxville

Jason Michael Crye
May 2013
DEDICATION

While pursuit of my doctoral degree has required much time and work from me, it has also required sacrifices from my family. In fact, my youngest daughter was born in August of 2007, the same month that I began pursuing this degree. She has only known me while I have been a graduate student! I owe my children much gratitude for understanding when I could not play due to studying, doing homework, or working on this dissertation. I hope that seeing me pursue this degree teaches them to never give up on their dreams and be committed to life-long learning. In addition to thanking my children, I also must thank my wife for supporting me in this endeavor. Since it was only a little over a year after finishing active duty with the U.S. Navy when I mentioned I wanted to go back to school, Libby initially was not very excited. However, it did not take her long to see how important this degree is to me, and since then, she has constantly been supporting and encouraging me. She has taken care of the children and our home when I could not, and again I owe her many thanks. Therefore, this dissertation is dedicated to my wife, Libby, and our children Mary Curtis, Gabriel, and Caroline.
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A research project of this scale is seldom the sole work of one individual. Indeed, I am indebted to many for their time, help, and support.

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anything she can to help me finish this degree. Seventh, Seth McConchie wrote the original proposal for this research (based on an idea from John Mihalczo) and convinced DOE that this research was worth funding. He is the principal investigator on the project and has managed much of the work. I appreciate him giving me the opportunity to work with him. Finally, Paul Hausladen came up with the idea for performing fission mapping reconstructions and provided the software that was used for the reconstructions discussed in Chapter 5. In addition to providing the software, he also has given me encouragement. When I first started working with time correlation measurements and imaging, I was a little overwhelmed. Paul told me I was like a squirrel jumping out in a major intersection and dodging cars coming from seemingly every direction. He stated that if you do not get run over, you will start to notice the cars travel between certain lines in the road, in certain directions, and at certain times corresponding to the color of the stop light. I have not forgotten this analogy. Since I have completed this dissertation, I do not think I have been run over. 😊

In addition to those at ORNL who are directly involved in the research, there are those who have provided support. In particular, my division director, Alan Icenhour, helped me craft a request for an educational sabbatical from ORNL, supported this request with upper management, and resolved several complex managerial issues related to the sabbatical. This sabbatical allowed me to finish my classwork and permitted my participation on this research project. So, I am grateful to both Alan and upper management for their support in pursuing this degree.
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Finally, I need to thank the U.S. Department of Energy National Nuclear Security Administration Office of Defense Nuclear Nonproliferation Research and Development for sponsoring this research.
ABSTRACT

The determination of the enrichment of uranium is required in many safeguards and security applications. Typical methods to determine the enrichment rely on detecting the 186 keV gamma ray emitted by uranium-235. In some applications the uranium is surrounded by external shields, and removal of the shields is undesirable. In these situations, methods relying on the detection of 186 keV gamma rays fail because these gamma rays are shielded easily.

This research presents a novel 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 plastic scintillation detectors following interrogation of the uranium with a deuterium-tritium neutron generator. The simulated time correlations that best match the measured time correlations are used to estimate the actual enrichment.

The method was demonstrated with measurements of a 93% enriched storage casting surrounded by different combinations of depleted uranium shields. For each combination, the fast neutron imaging techniques provided reasonable estimates of the known geometry and materials. Using the estimated geometry, the storage casting was modeled with several enrichments. The comparison of the measured time correlations to the predicted ones for each shielding combination clearly shows that the enrichment of
the casting is greater than 80%. By comparing the total doubles measured to the total doubles predicted from the simulations, the estimated enrichment of the casting is between 82% and 95% for the shielding combinations considered. Even though the worst estimate differs from the actual enrichment by 11%, the accuracy of the method is likely acceptable for many nonproliferation applications, including arms control and treaty verification where the goal may be simply to identify the presence of highly enriched uranium.
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2. Cf0001.PPI – Sample input file for PoliMiPPv6

3. Cf_32ImagingDets_8FissDets_16DtPix_2012-02-20.dsd.txt – Sample detector setup file
Uranium as mined from the earth is called natural uranium and has an isotopic composition of approximately 99.28% $^{238}\text{U}$, 0.71% $^{235}\text{U}$, and 0.006% $^{234}\text{U}$ [1]. Beginning with the Manhattan Project, there was a need for uranium materials that had a higher proportion, or enrichment, of the isotope $^{235}\text{U}$. The enrichment of uranium is defined as

$$x_e = \frac{m_{235}}{m_{238} + m_{235} + m_{234}},$$

where $m$ is the mass of each isotope in grams [2]. There is a wide range of enriched uranium material. Fuel where the $^{235}\text{U}$ content is greater than natural uranium but less than 20% is defined in federal regulations as Low Enriched Uranium (LEU) [3]. The fuel in most nuclear reactors is enriched to 3 to 5%. Fuel with a $^{235}\text{U}$ content greater than or equal 20% is defined in federal regulations as Highly Enriched Uranium (HEU) [3]. Some research reactors, naval reactors, and nuclear weapons use HEU. For example, the fuel in the High Flux Isotope Reactor at Oak Ridge National Laboratory (ORNL) is 93% enriched [4]. Uranium material that has a content of $^{235}\text{U}$ less than natural uranium is referred to as Depleted Uranium (DU).

Due to the high value of enriched uranium and to prevent proliferation, most countries have strict requirements controlling protection and accountability of the material. For example, before accepting responsibility for material shipped from another facility, a site generally should require confirmatory measurements of the material’s mass and enrichment. In most cases, the material is removed from its shipping container, and
the mass is measured with a scale. Typical methods to determine the enrichment rely on detecting low-energy photons. In some applications the uranium is surrounded by external shields, and removal of the shields is undesirable. For example, due to radiological control requirements, removal of the uranium metal from the storage containers often occurs in glove boxes, and there are times when glove boxes may be unavailable. In such situations, enrichment methods relying on the detection of the low-energy gamma rays may be degraded or even fail because the gamma rays are easily shielded.

While materials protection, control, and accounting requirements typically specify high accuracy for enrichment measurements, at least two applications may not need high accuracy. First, authorities assessing an object from a terrorist may only need to know if the object contains HEU to help determine if the object is a nuclear explosive device or a radiological dispersal device. Knowledge of the enrichment will help determine how to handle the device. Secondly, officials involved in verifying the agreements in arms control treaties may also only need a rough estimate of the enrichment. In both scenarios, the uranium metal is likely to be shielded.

This research involves developing a new method of estimating the enrichment of uranium metal when shielded by high-Z materials by combining fast neutron imaging techniques, time correlation measurements, and Monte Carlo simulations. In the first step of the method, a source of time and directionally tagged fast neutrons interrogate the object and an array of small and large plastic scintillation detectors measure the response during a short window following each interrogation. Next, tomographic
reconstruction algorithms are applied to the detector response to create an image that reveals the geometry and materials within the storage container. In the third step, the geometry and materials information is incorporated into Monte Carlo simulations. In each simulation, the uranium object identified in the fast neutron image is modeled with a different enrichment. Each simulation predicts the time distribution of coincidences that should be observed by the large scintillation detectors. Finally, the enrichment of the uranium is estimated by matching the measured time response to the closest prediction from the simulations.

Unlike gamma rays, neutrons easily penetrate high-Z shielding materials because these materials have relatively low neutron cross sections. Furthermore, calibration standards are not needed since any geometry and enrichment can be modeled with the Monte Carlo simulations. While the proposed method may not be as accurate as typical methods, it does allow inspection of unknown items of unknown enrichment in heavily shielded storage containers and is certainly adequate for nonproliferation applications.

**Justification and Original Contributions**

As discussed in the Chapter 2 literature review, typical methods that rely on the detection of low-energy photons to determine the enrichment of uranium metal fail when the metal is shielded by high-Z materials. Myers’s method based on delayed neutron die-away shows promise, but at the time of this research, it had only been tested with up to 1.6 cm of lead [5]. In addition, it is unclear how his method would work in the presence of fissionable shields.
This research is a novel approach to estimating the enrichment of uranium metal by combining fast neutron imaging, time correlation measurements, and Monte Carlo simulations. Using these three fields or techniques, the predicted enrichment of an 18 kg uranium storage casting when surrounded by varying layers of DU shielding was between 82% and 95%. The actual enrichment of the storage casting is 93%. With the possible exception of Myer’s method, other existing techniques likely could not provide estimates as accurate as these under the same shielded conditions. Furthermore, the time correlation measurements of the 93% enriched storage casting and a DU storage casting surrounded by varying layers of DU shielding are new and can serve as benchmarks for improving and testing techniques for modeling plastic scintillator detectors. In conclusion, the use of fast neutron imaging, time correlation measurements, and Monte Carlo simulations to determine the enrichment of uranium is new, has never been demonstrated before, and addresses a current weakness in existing methods.

Monte Carlo Simulations

Monte Carlo programs sample the Boltzmann transport equation and thus model how radiation moves throughout a volume. They can also predict neutron multiplication due to the presence of fissile or fissionable material. Lewis and Miller [6] provide an excellent derivation of the Boltzmann transport equation, so the derivation is not repeated here. The transport equation is
\[
\frac{1}{v} \frac{\partial \psi(\vec{r}, \hat{\Omega}, E, t)}{\partial t} + \hat{\Omega} \cdot \nabla \psi + \psi(\vec{r}, \hat{\Omega}, E, t) \sigma_i(\vec{r}, E)
\]

\[
= q_{\text{ex}}(\vec{r}, \hat{\Omega}, E, t)
\]

\[
+ \int_0^\infty dE' \int_{4\pi} d\Omega' \sigma_s(\vec{r}, E' \rightarrow E, \hat{\Omega}' \cdot \hat{\Omega}) \psi(\vec{r}, \hat{\Omega}', E', t)
\]

\[
+ \chi(E) \int_0^\infty dE' \nu(E') \sigma_f(\vec{r}, E') \int_{4\pi} \psi(\vec{r}, \hat{\Omega}', E, t) d\Omega',
\]

where \(v\) is the particle’s speed. The term \(\psi(\vec{r}, \hat{\Omega}, E, t) \ dV \ dE \ d\Omega\) represents the angular flux of particles in volume element \(dV\) at position \(\vec{r}\) traveling in a cone of directions \(d\Omega\) about \(\hat{\Omega}\) with energies between \(E\) and \(E + dE\) at time \(t\). \(\sigma_i\) is the total macroscopic cross section, and \(q_{\text{ex}}\) represents particles from external sources. The term \(\sigma_s(\vec{r}, E' \rightarrow E, \hat{\Omega}' \cdot \hat{\Omega}) \ dE \ d\Omega\) represents the scattering macroscopic cross section that gives the probability that particles at position \(\vec{r}\) scatter from energy \(E'\) to \(dE\) about \(E\) and from direction \(\hat{\Omega}'\) to \(d\Omega\) about \(\hat{\Omega}\). \(\chi(E) \ dE\) represents the probability that a fission neutron will have an energy \(dE\) about \(E\). \(\nu(E)\) is the mean number of neutrons produced from a fission caused by a neutron with energy \(E\). Finally, \(\sigma_f(\vec{r}, E)\) is the fission macroscopic cross section.

Seven independent variables comprise the phase space in the Boltzmann transport equation – three for position, two for direction, one for energy, and one for time. The equation has no analytical solution. However, there are several approximations or assumptions that are sometimes valid, such as diffusion theory or discretizing the angular variable, that allow solving for the flux. Every method of computing the flux with the transport equation requires input of geometry, materials,
cross-sections, boundary conditions, external sources, and an initial value of the flux if the
time-dependent form of the transport equation is used.

Monte Carlo programs sample the transport equation by following the “lives” of particles created from the user-specified source and recording when they cross specified boundaries or other such tallies. For example, suppose that a particle is created in a simulation from the user-specified source. The programs then sample distributions using random numbers to determine the particle’s exact position in the source, its direction, and its energy. Then, the programs calculate how far the particle will travel in the material before having an interaction by sampling a distribution based on the total cross section. If the particle does not leave the material before having an interaction, the programs determine the type of reaction by sampling a distribution based on the fission, scattering, and absorption cross sections. If the reaction is fission, the programs track each particle created by the fission. If the reaction is scattering, the programs determine the new direction and energy by sampling a distribution based on the double differential cross section. If the reaction is absorption without creation of any secondary particles, the life of the particle or history terminates. This process continues until the particle escapes the user-specified geometry, exceeds a user-specified cutoff, or is absorbed. The programs then start the process over again with a new particle and history until a specified number of particles have been simulated. Finally, after the program finishes, the user evaluates the tally to determine if the result is statistically significant. If the result is not statistically significant, the process is repeated until the desired statistical significance is obtained.
**Time Correlation Measurements**

In the context of this dissertation, the term time correlation measurements simply refers to measurements where the detection time of radiation is recorded and analyzed for any correlations. Time correlation measurements are ideally suited for samples that include fissile or fissionable material since fission typically results in the simultaneous creation of more than one prompt neutron. The number of prompt neutrons produced in a fission, $\nu$, follows a Gaussian distribution as shown by Terrell [7]. Therefore, the probability that $\nu$ prompt neutrons are produced from a fission event is given by

$$P(\nu) = \frac{1}{\sqrt{2\pi}\sigma^2} e^{-\frac{(\nu-\nu)^2}{2\sigma^2}},$$  \hspace{1cm} 1-2

where $\sigma$ is the width of the distribution. Terrell has shown that a $\sigma$ value of 1.08 is appropriate for several isotopes including induced fission of $^{235}$U. However, Terrell’s data suggests that a higher value of $\sigma$ may be needed for induced fission of $^{235}$U with neutrons having energies above 1.25 MeV. Nevertheless, to illustrate use of Equation 1-2, Figure 1-1 shows the distributions for induced fission of $^{235}$U with a thermal neutron and a 14 MeV neutron using a $\sigma$ value of 1.08. Such distributions are often called multiplicity distributions. Not all of the fission neutrons are available for detection. Some are captured in the sample or surrounding materials and, in a multiplying medium, others induce more fission. Nevertheless, the neutrons that are detected typically do not arrive at the detectors at the same time. Depending upon where the fission occurs in relation to the detectors, each fission neutron may have to travel a different distance to reach one of the detectors. Furthermore, the fission neutrons do not travel at the same speed.
Even though the fission neutrons are not detected at the same time, there are correlations between the detection times.

Two primary time distributions are used in the simulations and measurements in this research, and both are based on the detection time of an interrogating particle. The first time correlation is the singles distribution. It is a histogram that shows how the detection times of the particles vary with respect to the creation time of the interrogating particle. Particles from the background are detected at random times with respect to the interrogating particle, but particles resulting from interactions such as induced fission have a specific range of detection times. The second distribution is the doubles distribution. It is a histogram that shows the difference in detection times between all possible pairs in a selected time window. The window is selected to include the time
when prompt neutrons are expected. If fissionable material is present, then more pairs are detected. Mihalczo describes the doubles distribution as the cross correlation function between detectors and states that it is essentially the same as the Rossi-α distribution from reactor noise analysis [8].

The time correlation measurements in this research are related to conventional neutron coincidence counting as described by Ensslin [9] and multiplicity counting as described by Ensslin et al. [10]. Both coincidence and multiplicity counters often detect neutrons with rings of $^3$He tubes contained within polyethylene. The polyethylene is used to thermalize the fission neutrons because $^3$He detectors primarily detect thermal neutrons. Therefore, the counters operate on a time scale of microseconds to allow time for thermalization. In contrast, time correlation measurements in this research typically employ fast plastic or liquid scintillation detectors and operate on the time-scale of nanoseconds, which is the time-scale of the fission chain multiplication process for subcritical configurations of uranium metal. Next, coincidence and multiplicity counters do not record the detection times of all the particles. Therefore, they are not associated with singles and doubles distributions as previously described. Coincidence counters use specialized circuits called shift registers to record the number of neutron pairs that have a small time difference and the number that have a large time difference. The number of pairs with a large time difference are called the accidental coincidences. The number of pairs with a small time difference that are in excess of accidental coincidences are called real coincidences since they are due to fission neutrons. Multiplicity counters use a
modified shift register circuit that records the multiplicity distribution. In other words, the multiplicity counters measure the number of times \( \pi \) neutrons are detected.

**Fast Neutron Imaging**

Due to the prevalent use of computed tomography (CT) scans and positron emission tomography (PET) scans in medicine, the concept of rotating an array of detectors around an object and creating an image that shows the object’s internals is generally familiar. Kak and Slaney describe the technique implemented in most CT scanners [11], and Mullens describes the fast neutron imaging technique used in this research [12]. The primary difference between the techniques is that CT scanners use the attenuation of x-rays through the body to create an image, whereas this fast neutron imaging uses the attenuation of 14 MeV neutrons from a deuterium-tritium (DT) neutron generator.

All tomographic techniques begin with measuring a ray integral, which is mathematically defined as the integral of a function along a line. To illustrate its use in imaging, consider a monoenergetic beam of particles passing through an object. Let \( I_o \) represent the number of particles in the beam prior to entering the object and \( I \) be the number after passing through the object. Then, it follows that

\[
I = I_0 e^{-\int \sigma_r(s) ds},
\]

where \( s \) is the distance travelled. Notice that the cross section in this case is a function of the distance travelled to allow for different materials in the object. Solving for the integral gives
\[
\int_{ray} \sigma_t(s) \, ds = \ln \left( \frac{I_0}{I} \right).
\]

The variable \( I_0 \) is determined from a measurement where the object is not present, and the variable \( I \) is determined from a measurement where the object is present. Therefore, by measuring \( I_0 \) and \( I \), the integral of the cross section along the line is known.

Two types of beam and detector arrangements are used in tomography. The most common type is a fan beam arrangement, which is shown in Figure 1-2 on the right. In a fan beam arrangement, the beams originate from a single source, and the detectors are typically positioned in an arc such that the source to detector distance is a constant for all the detectors. Note in Figure 1-2 that each arrow represents a ray integral measured with a detector. Each set of ray integrals forms a projection, and the projections are shown as line graphs in Figure 1-2. The second type of arrangement is a parallel beam, which is shown in Figure 1-2 on the left. The parallel beam is simpler to understand but often more complex to implement in practice. Regardless, Kak and Slaney show that the projections measured with a fan beam can be mathematically converted to parallel projections [11].

There are several algorithms that can perform the image reconstruction from the projections. Lalush and Wernick discuss several iterative reconstructions methods, such as the algebraic reconstruction technique and the maximum-likelihood expectation maximization algorithm [13], but the most common reconstruction technique is filtered backprojection as described by Kak and Slaney [11].
The first step in filtered backprojection as described by Kak and Slaney is to convert the projections to the frequency domain. For a parallel projection, the Fourier Slice theorem states that the Fourier transform of a projection of the function $f(x, y)$ measured at an angle $\theta$ from the abscissa axis gives the values of the function $F(u, v)$ in the frequency domain along a line measured at the same angle from the abscissa axis. Figure 1-3 illustrates the Fourier Slice Theorem. Using the nomenclature from Kak and Slaney, let $P_\theta(t)$ represent a projection at an angle $\theta$. For each projection, the variable $t$ is the distance in the direction of $\theta$ from each ray to the ray that passes through the origin in the space domain.
The Fourier transform of $P_\theta(t)$ is given by the equation

$$S_\theta(\omega) = \int_{-\infty}^{\infty} P_\theta(t) e^{-j2\pi\omega t} dt,$$

where $\omega$ is the frequency and $j$ is the imaginary number $\sqrt{-1}$. In Figure 1-3, the directions $u$ and $v$ are related to the frequency by the equations $u = \omega \cdot \cos(\theta)$ and $v = \omega \cdot \sin(\theta)$. If an infinite number of projections could be measured, the frequency domain would be completely known. However, since only a finite number of projections can be measured, the frequency domain is populated as shown in Figure 1-4. Notice that there are more points in the center than in regions away from the center. Therefore, the center region in the reconstructed image is more accurate due to the higher sample density. Without any filtering, the reconstructed image would also have a starburst appearance due to the lower sample density in regions away from the center.
To reduce the starburst appearance and reduce blurring in the image, the frequency domain projections, $S_\theta(\omega)$, are multiplied by the ramp filter $|\omega|$. The ramp filter increases the weight of the points away from the center such that the weighted sample density is more uniform.

After applying the ramp filter, the next step in the filtered backprojection algorithm is taking the inverse Fourier transform of each projection such that

$$Q_\theta(t) = \int_{-\infty}^{\infty} S_\theta(\omega) |\omega| e^{j2\pi \omega t} d\omega,$$

where $Q_\theta(t)$ is the filtered projection. Note that it is possible to perform the filtering in the spatial domain and avoid the Fourier transformations.

Finally, the image is created by summing the contribution of each filtered projection to each point in the space domain. This process is called backprojection. In practice, a grid of points is selected in the space domain. For each projection, there is a
value of $t$ that passes through each $(x, y)$ point on the grid. Therefore, the reconstructed image is found by

$$\hat{f}(x, y) = \frac{\pi}{K} \sum_{i=1}^{K} Q_{\theta_i}(x \cos(\theta_i) + y \sin(\theta_i)),$$

where $K$ is the number of projections between 0 and 90°. Recall that for our imaging example, the original function used in the ray integral is the cross section. So, the reconstructed image shows the cross section throughout the space domain.
CHAPTER 2

PRIOR WORK

Several techniques involving both gamma rays and neutrons have been used to determine the enrichment of uranium.

Gamma Spectroscopy

The use of gamma spectroscopy to determine the enrichment of uranium first appeared in 1968 when James Russell received a patent [14]. His method, as well as most subsequent extensions and refinements, relies on the detection of the 186 keV gamma ray emitted by $^{235}$U. Russell connects a scintillation detector to two single channel analyzers. One single channel analyzer records the counts at 186 keV. The other records the counts at 250 keV to give a measure of background or Compton continuum contribution at a position near 186 keV. The Compton continuum from the higher energy gamma rays associated with $^{238}$U is in both single channel analyzers. Therefore, the enrichment is determined by

$$x_e \propto \frac{SCA_{186\,keV} - SCA_{250\,keV}}{SCA_{250\,keV}}, \quad 2-1$$

where $x_e$ is the enrichment and $SCA$ is the counts recorded from the single channel analyzer.

In 1970, Reilly et al. developed the enrichment meter technique, which is the technique most widely used to determine uranium enrichment [15, 16]. With this technique, the sample does not have to be the same size as the standards used for
calibration. To limit the detector’s field of view, a collimator is placed between the detector and sample. Next, the technique specifies that the thickness of the sample near the opening in the collimator must be greater than approximately seven mean free paths of the 186 keV gamma ray so that the sample is opaque to 186 keV gamma rays arising deeper in the object. With a collimator and sufficiently thick sample, there is only a small volume of the sample that is visible to the detector. Therefore, the actual size and geometry of the sample is not important provided this infinite-thickness criterion is met. The technique assumes that the enrichment in the sample is uniform. Samples having different enrichments are needed for calibration, and variances in enrichment below the gamma-transparent layer will be missed.

In 1975, Harry et al. presented what became known as the peak ratio technique [17]. This technique offers the advantage that neither the infinite thickness criterion nor calibration standards are needed. Since the height of a photpeak in a gamma spectrum is proportional to the activity of the isotope that emitted the gamma radiation, the ratio of the heights of specific photopeaks is used to determine the enrichment. As before, the photpeak at 186 keV is due to $^{235}$U. There are several photpeaks that can be used to determine the $^{238}$U activity. $^{234m}$Pa is a daughter of $^{238}$U and emits gamma rays with energies of 743, 766, 786, and 1001 keV. If $^{234m}$Pa is in secular equilibrium with $^{238}$U, then the height of any of these photpeaks can be used with the height of the 186 keV peak to determine the enrichment. Since there is a large energy difference between the 186 keV gamma ray associated with $^{235}$U and the gamma rays associated with $^{238}$U, a correction term must be added to account for the different detection efficiencies [2].
In the 1980s, Gunnink developed a peak fitting technique for determining the isotopic abundances of plutonium and applied it in the Multi-Group Analysis (MGA) code [18]. In 1994, he used the same peak fitting technique to determine the enrichment of uranium and developed the MGAU code [19]. For uranium, Gunnink analyzes a high-resolution spectrum from a germanium detector in the energy range of 90 to 120 keV. In this energy range, both $^{235}$U and $^{238}$U and their daughters emit gamma rays and x-rays, but the energies of the different photons are close and overlap. Therefore, the MGAU code applies a response function to unfold the overlapping peaks. Essentially, the code computes a spectrum and iterates on the amount of $^{235}$U and $^{238}$U present until the computed spectrum matches the measured spectrum. A calibration is not required, but there must be secular equilibrium between the $^{235}$U and $^{238}$U parents and their daughters [19]. Based on the same principle of unfolding the peaks in the 90 to 120 keV region, Morel developed the URADOS method in 1998 [20].

In addition to applying the peak fitting technique to the gammas and x-rays in the 90 to 120 keV range, Gunnink applied it to NaI spectra in the energy range of 130 to 290 keV and developed the computer code NaI Gamma Enrichment Measurements (NaIGEM) [21]. The method computes a 186 keV peak and iterates on the amount of $^{235}$U present until the computed spectrum matches the measured spectrum. Since the method is based on the 186 keV gamma ray from $^{235}$U, the method is similar to the enrichment meter method.

Another gamma spectroscopic method is implemented in the Blend Down Monitoring System (BDMS). In 1993, the US and Russian Federation agreed to
downblend the HEU from Russian nuclear weapons into LEU that is used as fuel by US nuclear power plants. To provide transparency for the downblending process, Los Alamos National Laboratory and ORNL developed the BDMS with the requirement that no penetrations are allowed into the pipes carrying the UF$_6$ gas. The enrichment meter in the BDMS includes a $^{57}$Co source on one side of a pipe and a NaI detector on the other side. Once again, the intensity of the 186 keV gamma ray in the spectrum observed by the NaI detector is used to determine the $^{235}$U content. To determine the total uranium content, the attenuation of the 122 keV gamma ray from $^{57}$Co is used. Of course, corrections are made to account for the attenuation of the pipe walls. [22]

All the gamma spectroscopic methods involve examining photons with 186 keV of energy or less. The photons are easily shielded by any high-Z materials between the detector and the sample. With only a small thickness of shielding, the methods will fail. Similarly, due to the self-shielding of uranium, the methods only provide a surface measurement. The necessity of detecting low-energy photons limits the application of gamma spectroscopic methods.

**Passive Neutron Detection**

Neutrons have the advantage of being able to penetrate deep within high-Z materials.

Table 2-1 shows decay data for $^{235}$U and $^{238}$U [23]. The specific activity, $A_s$, is computed by

$$A_s = \frac{\ln 2}{t_{\frac{1}{2}}} \cdot \frac{N_A}{A_w}$$

2-2
where \( t_{1/2} \) is the half-life, \( N_A \) is Avogadro’s number, and \( A_w \) is the atomic weight. The average neutron emission rate from spontaneous fission (neglecting multiplication) is calculated by

\[
E_R = A_s \cdot br_{sf} \cdot \bar{\nu},
\]

where \( br_{sf} \) is the branching fraction for spontaneous fission and \( \bar{\nu} \) is the average spontaneous fission multiplicity. As shown, the neutron spontaneous fission emission rate in \(^{235}\text{U}\) is essentially negligible when compared to the rate from \(^{238}\text{U}\). Therefore, enrichment of uranium cannot be determined by simply counting the number of neutrons emitted from spontaneous fission.

Even though enrichment cannot be determined from spontaneous fission, total neutron counting has been used to determine the enrichment of UF\(_6\) [24]. When the uranium isotopes do not decay by spontaneous fission, they can decay by \( \alpha \) emission.

### Table 2-1. Decay data for uranium

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>( t_{1/2} ) (yr)</th>
<th>Specific Activity (disintegrations per second per gram)</th>
<th>Spontaneous Fission Branching Fraction</th>
<th>Spontaneous Fission Multiplicity, ( \bar{\nu} ) (average number of neutrons produced per fission)</th>
<th>Neutrons/(s-gram)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{235}\text{U})</td>
<td>( 7.04 \times 10^8 )</td>
<td>( 8.00 \times 10^4 )</td>
<td>( 7.2 \times 10^{-11} )</td>
<td>1.87</td>
<td>( 1 \times 10^{-5} )</td>
</tr>
<tr>
<td>(^{238}\text{U})</td>
<td>( 4.47 \times 10^9 )</td>
<td>( 1.24 \times 10^4 )</td>
<td>( 5.5 \times 10^{-5} )</td>
<td>2.00</td>
<td>1.37</td>
</tr>
</tbody>
</table>
In fact, $^{234}\text{U}$ is the primary uranium isotope that emits $\alpha$ particles due to its relatively short half-life ($2.45 \times 10^5$ years). Since the atomic weight of $^{234}\text{U}$ is close to $^{235}\text{U}$, the concentration of $^{234}\text{U}$ is related to the $^{235}\text{U}$ enrichment. For the gaseous diffusion enrichment process, Smith [2] reports that the ratio of $^{235}\text{U}$ to $^{234}\text{U}$ can vary by a factor of 4 from the range of depleted uranium to highly enriched uranium. The $\alpha$ particle has a short range, but if certain low Z atoms such as $^{18}\text{O}$ and $^{19}\text{F}$ are nearby, the $\alpha$ particle can cause an ($\alpha$,n) reaction. Stewart reports for 10 kg of UF$_6$, 20% and 98% enrichments generate 5,457 and 60,713 neutrons/second, respectively, due to ($\alpha$,n) reactions [24]. Therefore, a detector can be calibrated to provide the enrichment of UF$_6$ based on the neutron count rate. This method will not work for uranium metal.

**Active Neutron Interrogation**

Since the spontaneous fission rate of uranium is not sufficient for determining the enrichment, uranium samples are often bombarded with external neutrons to induce fission. Californium-252 and neutron generators are used to provide the external neutrons. Active neutron interrogation techniques take advantage of the fact that $^{235}\text{U}$ always has a higher fission cross section than $^{238}\text{U}$, as shown in Figure 2-1 [23]. Even with a 14 MeV neutron from a deuterium – tritium neutron generator, the $^{235}\text{U}$ fission cross section is almost twice as much as the $^{238}\text{U}$ cross section. Therefore, samples with higher enrichments will fission more and create more neutrons.
**Delayed Neutron Die-Away**

Myers, Gooulding, and Hollas present a method based on re-interrogation with delayed neutrons [5]. After fission, several prompt neutrons and γ-rays are immediately emitted. Some of the fission fragments, called delayed neutron precursors, will beta-decay into an isotope that emits a delayed neutron. Delayed neutrons account for less than 1% of the all the neutrons emitted from a fission event [25]. The time between the fission event and the emission of the delayed neutron is controlled by the beta-decay half-life of the precursor. Although there are many possible delayed neutron precursors, they are typically categorized into six groups based on their half-life. Each group’s
characteristic half-life ranges from \(~0.2s\) to \(~55s\). In the re-interrogation method, Myers bombards samples of uranium with neutrons from a DT generator until the delayed neutron precursor concentrations reach steady state. Then, the DT generator is turned off and delayed neutrons re-interrogate the sample and cause fission. The neutron count rate is measured with 48 \(^3\)He detectors. The neutron count rate decreases with time, and the decrease or die-away is dependent on the sample enrichment and multiplication.

To extract the enrichment, Myers begins with an equation provided by Li [26] that describes the delayed neutron count rate for a small sample after irradiation by fission-inducing particles. Myers modifies the equation to account for multiplication and a sample with two fissionable isotopes. After some algebra and substitution into the basic equation for enrichment, the enrichment is expressed as

\[
x_e = \frac{\nu_{d238} \cdot \sigma_{f238} \cdot B_{235} \cdot M_{235}}{\nu_{d238} \cdot \sigma_{f238} \cdot B_{235} \cdot M_{235} + \nu_{d235} \cdot \sigma_{f235} \cdot B_{238} \cdot M_{238}},
\]

where \(\nu_d\) = average number of delayed neutrons emitted per fission, \(\sigma_f\) = fission cross section (barns), and \(M\) = atomic mass number (g/mol). The coefficients \(B_{235}\) and \(B_{238}\) are obtained by fitting the measured count rate data as a function of time to the equation

\[
C_T(t) = B_{235} \cdot \sum_{i=1}^{6} \beta_{235i} e^{-\lambda_{235i} t} + B_{238} \cdot \sum_{i=1}^{6} \beta_{238i} e^{-\lambda_{238i} t},
\]

where \(C_T(t)\) = the measured count rate at time \(t\), \(\beta\) = fraction of delayed neutrons emitted in group \(i\), and \(\lambda\) is the decay constant of the delayed neutrons in group \(i\). Note that \(\beta_i\) and \(\lambda_i\) are different for each isotope of uranium.

With this method of total counting based on delayed neutrons, Myers estimated the enrichment of six uranium oxide samples that had approximately the same mass but
differing enrichments. He also estimated the enrichment of HEU and DU metal samples including when shielded by 1.6 cm of lead. In all the cases, the estimated enrichment was within 20 weight percent of the actual samples. In most cases, the estimates differed by no more than 10 weight percent. With better fitting of the $B_{235}$ and $B_{238}$ coefficients, Myers considers more accurate enrichment estimates are possible [5].

**Time Correlations**

Mattingly et al. present a method of determining enrichment based on active neutron interrogation and coincidence counting with the Nuclear Materials Identification System (NMIS) [27]. The method was used in 2000 to verify the mass and enrichment of several (~500) HEU metal items in their bird cages\(^1\) and successfully resolve a Department of Energy finding at the Y-12 National Security Complex. In the application, time-tagged neutrons from a $^{252}$Cf source interrogate the uranium metal, and the gamma rays and neutrons emitted from induced fission are detected with fast plastic scintillation detectors. Since samples with higher enrichments and mass have more fission, the time distribution of detected counts following creation of the interrogating neutron is different for samples having different enrichments and/or masses.

To extract the enrichment and mass from the measured time distribution, Mattingly developed empirical models that relate the zero and first factorial moment of the time distribution to the enrichment and mass. The empirical models are based on Monte Carlo simulations. In the simulations, Mattingly computes the time distribution

\(^1\) A bird cage is an open container at Y-12 that keeps HEU metal items separated for criticality safety.
and associated moments for cases where the enrichment and mass vary over the ranges expected from the items. The results give a non-linear unique calibration surface where the enrichment and mass are predicted based on the two moments. To validate the calibration surface, the enrichment and mass were predicted for ten items. Afterwards, the ten items were removed from their storage containers, and the enrichment and mass were measured by typical \( \gamma \)-ray spectroscopy methods. Comparison of the predictions to the measurements showed that the calibration surface had a bias due to the simulations. Once the bias was incorporated into the surface, the remaining items were interrogated, and the method was used to predict the mass and enrichment of the items. Even though none of the predicted mass and enrichments deviated from the declared values by more than 5%, Mattingly states the method in current form is not generally applicable to estimate the enrichment of all HEU metal items [27].
CHAPTER 3

METHOD

In this research, the method used to determine the enrichment of a shielded object involves fast neutron imaging, time correlation measurements, and simulations. To perform the fast neutron imaging and time correlation measurements, the Nuclear Materials Identification System (NMIS) [8] developed by Oak Ridge National Laboratory is used. NMIS has evolved over time, and Grogan provides a good history of its development [28]. NMIS uses a DT generator with an associated particle detector to provide a source of fast neutrons for interrogating an object. On the opposite side of the DT generator, NMIS has an array of 32 small plastic detectors in a fan beam arrangement. The small detectors are called imaging detectors since they are used to create a tomographic image. Next, NMIS includes eight large plastic detectors that are used to detect the induced fission radiation. The signals recorded by these fission radiation detectors form the basis for the time correlations used for this enrichment determination. Including the associated particle detectors, there are a total of 56 detectors in the existing version of NMIS.

To control the position of the DT generator, imaging detectors, and the target for the measurements, NMIS uses three motors. The first motor controls the height of the imaging detectors and DT generator. By raising and lowering the DT generator and imaging detectors, NMIS can interrogate an object at differing heights. The second motor controls a turntable. If the object is not cylindrically symmetric, the object must be
incrementally rotated to obtain many projections for the tomographic images. If the object is cylindrically symmetric, the turntable is not needed since the counts recorded at each rotation will be the same. Finally, the third motor controls the horizontal position of the fan beam of imaging detectors.

Typically when interrogating a cylindrically symmetric object with NMIS, several short measurements are made at different heights to show the vertical profile of the object. In addition, tomographic images at these heights can be created to show how the interior geometry changes with height. After the short measurements, a longer measurement is made at the mid-height of the object to obtain good statistics from the data recorded by the fission radiation detectors.

After the measurements are complete, an analyst must estimate the geometry and materials in the object based on the tomographic images. The analyst then builds models for Monte Carlo simulations using the estimated geometry and materials. If the object contains uranium, the analyst models the parts assumed to be enriched with several enrichments. For each enrichment, the simulations predict the time correlations that should be observed by the fission radiation detectors. Finally, the analyst estimates the enrichment by determining which simulated time correlations best match the measured time correlations.

To test this method of determining the enrichment of shielded uranium, time correlation measurements are made of unclassified annular uranium storage castings used at the Nuclear Detection and Sensor Test Center of the Y-12 National Security Complex in Oak Ridge, TN. The dimensions of the castings are given in Table 3-1, and
Table 3-1. Dimensions of annular 161 storage castings

<p>| | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Height (in)</td>
<td>6</td>
</tr>
<tr>
<td>Inside Radius (in)</td>
<td>1.75</td>
</tr>
<tr>
<td>Outside Radius (in)</td>
<td>2.5</td>
</tr>
<tr>
<td>Thickness (in)</td>
<td>0.75</td>
</tr>
</tbody>
</table>

the enrichments include DU and 93%. To shield the castings, up to three layers of DU, each about ½ in. thick, are placed around the castings. Finally, the measured time correlations are compared to the simulations.

The remainder of this chapter discusses the equipment used for the experiments and the Monte-Carlo code used for the simulations.

**Neutron Generator**

A Thermo Scientific API 120 neutron generator provides the source of neutrons by the reaction

\[
\frac{2}{1}H + \frac{3}{1}H \rightarrow \frac{4}{2}\alpha + \frac{1}{0}n .
\]

Conservation of momentum and energy shows that the neutron has a kinetic energy of 14 MeV and travels approximately 180° from the α particle. As shown in Figure 3-1, the neutron generator is primarily comprised of a rectangular box and a sealed cylindrical tube. The rectangular box contains the electronics for the generator. The sealed cylindrical tube contains the high-voltage supply, ion source, target, and a fiber-optic faceplate. The fiber-optic faceplate includes a thin layer of an inorganic scintillator (Yttrium Aluminum Perovskite) mechanically coupled to the inner surface on the inside of the generator to produce light whenever an α particle is detected. The API 120 is
Figure 3-1. Drawings of the Thermo API 120 neutron generator [29]

capable a producing up to $1 \times 10^8$ neutrons per second [30]. Oak Ridge National Laboratory (ORNL) provided Thermo Scientific the scintillator in the alpha detector of the generator. While earlier versions of the generator produce $1 \times 10^8$ neutrons per second, more recent ones produce $4 \times 10^7$ neutrons per second [30].

Mounted to the outside of fiber-optic faceplate of the neutron generator is a Hamamatsu H9500 position sensitive photomultiplier tube (PMT). As shown in Figure 3-2, the PMT contains a 16 x 16 grid of pixels, but only one row is used in this application. The dimension of each pixel of the PMT is 2.8 x 2.8 mm. A pixelated light
guide is placed between the PMT and the fiber-optic faceplate of the neutron generator to channel the light from the scintillator into the pixels of the PMT. The PMT is enclosed in an Al housing, which has an opening for a bias voltage cable and the 16 signal cables from the pixels. Since the α particle and neutron are created simultaneously and travel in almost opposite directions, the API120 neutron generator and H9500 PMT provide a source of time and directionally tagged neutrons.

**Imaging Detectors**

The image detector assembly is an array of 32 plastic scintillator detectors in a fan beam arrangement, as shown in Figure 3-3 and Figure 3-4. The detectors are manufactured by Scionix, and the dimension of each detector is 2.54 x 2.54 x 10.16 cm. The distance from the DT generator and each image detector is 110 cm. The fan beam on the NMIS is attached to a motor that moves the fan beam along the arc in
Figure 3-3. Arrangement of DT generator and imaging detectors
The DT generator is rotated 4.3° to center the neutrons on the fanbeam.

Figure 3-4. Picture of imaging detectors from behind neutron generator
increments that can vary. For these measurements, the increments are 1/4 the width of
the imaging detectors. Therefore, there are 128 measurement locations for each
projection. This process of moving the fanbeam in small increments to obtain more
measurements in each projection significantly improves the resolution of the
tomographic image.

Since the $\alpha$ particle and neutron from a DT reaction travel approximately 180°
apart, detections in each $\alpha$ pixel are strongly correlated with certain imaging detector
positions, as shown in Figure 3-5. The data Figure 3-5 are from a void measurement, and
the figure shows how neutrons detected at each image detector position are related to
each $\alpha$ pixel. As expected based on the locations shown in Figure 3-3,

![Figure 3-5. Correlation of $\alpha$ pixels with imaging detector positions based on void measurement](image-url)
most of the $\alpha$ counts in pixel 1 are associated with neutron counts in the imaging detectors 1-6 (detector positions 1-24).

**Fission Radiation Detectors**

Similar to the imaging detectors, the fission radiation detectors are also fast plastic scintillator detectors manufactured by Scionix. Each detector has the dimensions of $27 \times 27 \times 9.5$ cm. Since plastic scintillation detectors are sensitive to both neutrons and gammas, the front face of the detector includes a layer of Pb that is ¼ in. thick to prevent some of the low-energy gammas from reaching the detector.

The detectors are arranged on an aluminum stand as shown in Figure 3-6. The stand is designed to give each detector the same solid angle to a target placed at the center of the stand. The stand has two plates that support the detectors. The inside of each plate forms an arc with a radius of 48.25 cm from the center axis. The top of the detectors on the bottom row is 55.4 cm above the floor, and the bottom of the detectors on the top row is 85.5 cm above the floor. Hence, the mid-height where targets are centered is 70.5 cm above the floor. The distance from center axis at 70.5 cm above the floor to the center of the front of each detector is approximately 58 cm.
Data Acquisition System

To acquire the detector signals, NMIS uses two custom-made boards that each fit into a Peripheral Component Interconnect (PCI) slot in a computer. Mihalczo describes the details of the boards in reference [8]. Each board can receive five channels of data, and sample each channel at rates up to 1 GHz. Each board also has its own internal processor and clock, and the clocks must be synchronized periodically to minimize timing differences between the processor boards. To do the synchronization, both boards must receive an input signal that is exactly the same and whose pulses arrive at exactly the same time. The channel used for synchronization must contain only one input signal. Since two of the 10 channels available are used for synchronization, there are eight channels remaining for the detector signals. However, these remaining channels can
each receive more than one input signal. To associate the signals on a particular channel with a particular detector, the signals, which are logic pulses, have differing pulse widths.

**Routing of Signals to Data Acquisition System**

The signals from all the detectors are routed to the data acquisition system in a similar manner. After leaving a PMT, the signal is sent to a constant fraction discriminator (CFD). CFDs are often used in fast timing measurements to ensure that both large and small detector pulses start at the same time and thus reduce the time jitter from low and high pulses crossing the CFD threshold at different times. The CFDs also serve as lower level discriminators and convert each input linear pulse into a logic timing pulse. Since the computer used for data acquisition only has eight effective input channels for 56 detectors, the width of the logic pulse is set such that the combination of the pulse width and the input channel can uniquely identify which detector the signal came from. The CFDs used in this experiment are either the ORTEC 935 Quad CFD or the ORTEC CF8000 Octal CFD. After leaving the CFDs, the signals are sent to Phillips Scientific Model 757 Mixed Logic units, which are herein referred to as mixers. The mixed logic units combine up to 16 input signals into one output signal while preserving the pulse widths. Finally, the signals go to the NMIS input boards.

The wiring arrangement for the $\alpha$ pixels on the DT generator is shown in Figure 3-7. All of the $\alpha$ pixels use the same bias voltage. From the PMT, the signals are input into the ORTEC 935 Quad CFDs. Each model 935 CFD can condition up to four signals.
After leaving the CFDs, the signals are input into a mixer. Notice that the first mixer combines the signals from all the odd-numbered pixels. These signals are then sent to input channel 1 on the first NMIS input board. The second mixer combines the signals from all the even-numbered pixels. These later signals are sent to input channel 6 (the first input channel on the second board).

In addition to the routing for the signals from the α pixels, the wiring for the board synchronization signals is also shown on Figure 3-7. Typically, 50 mV noise pulses from a pre-amplifier for a Cf source are used for the synchronization signal. Before, being sent to the input boards, the pulses are routed to an ORTEC 935 CFD. Since the boards must receive the logical timing pulses from the CFD at the same time, care must be taken to ensure that the cables from the CFD to the boards are exactly the same length for proper synchronization.
The wiring for the image detectors is shown in Figure 3-8. The high voltage is supplied by a CAEN model N472 module. This module has four outputs which can be individually adjusted. Each output supplies bias voltage to eight imaging detectors. To conserve space in the Nuclear Instrument Module (NIM) bins, 4 ORTEC CF8000 CFD modules are used. Each CF8000 module can receive eight input signals. Even though the CF8000 module allows adjusting the threshold for each input signal, all output signals have the same pulse width. Therefore, each output signal from a CF8000 CFD must be sent to a different mixer and input channel on the NMIS boards. For example, the input to board 1 channel 2 has the signals from imaging detectors 1, 7, 13, 19, 25, and 31 since each signal has a different pulse width.

As shown in Figure 3-9, the signal routing for the fission radiation detectors is also similar to that for the $\alpha$ pixels and the imaging detectors with a few exceptions. First, each fission radiation detector has its own high-voltage supply and CFD to allow individual adjustment of the bias voltage and threshold. Next, the signals after leaving the CFDs are sent to a Phillips Scientific Model 794 Gate and Delay Generator NIM module. Finally, since there are no free input channels on the NMIS board, the signals are routed to mixers used primarily for the $\alpha$ pixels or imaging detectors.

The gate and delay generator inserts approximately a 300 ns delay for each signal. The fission radiation detectors are large and have a high count rate. Since some of the signals from the fission radiation detectors share the same input channel with the alpha pixels, the 300 ns delay ensures that the signals from the $\alpha$ pixels arrive and are processed by the NMIS input boards first. The data acquisition system allows
Figure 3-8. Wiring for the imaging detectors
adjustment of each signal’s time skew to ensure that the timing is correct. The adjustment of the time skews is discussed later.

**NMIS Data Acquisition and Analysis Software**

Chiang *et al.* discuss the two programs associated with controlling NMIS and analyzing the measurements [32]. The first was written by Mullens and is named Data Acquisition and User Interface (DAUI). DAUI is primarily used for operating NMIS during a measurement, calculating the time correlations, and recording the list mode data. Before a measurement, DAUI reads a file that specifies details for the specific detectors involved
in the measurement. For example, this file contains the channel number and pulse width range for each detector. An example of the file is shown and discussed in Appendix C.

DAUI also controls the three NMIS motors and allows the user to specify the details needed for a measurement such as the heights, sampling positions for the imaging detectors, rotations, and the time duration of the measurement. During a measurement, DAUI computes the time correlations both in time and frequency domains in real time. Finally, DAUI has the ability to load a void measurement ($I_0$ in Equation 1-4) and an object measurement ($I$ in Equation 1-4) and to create a tomographic image using filtered backprojection.

The second program is named Interactive Data Analysis Software (IDAS). It was written by Mullens and Blakeman. IDAS reads the binary file written by DAUI that contains the measured time correlations. Mihalczo discusses each of the correlations in detail in reference [8]. For example, IDAS can plot the cross-correlation of the signals from two detectors. If the first detector is an alpha pixel or the Cf ion chamber, the cross correlation is the singles distribution discussed in Chapter 1. IDAS can also plot the cross spectral power density between two detectors, which is the same as the fast Fourier transform of the cross correlation function. Finally, IDAS uses data from time-of-flight measurements in air with a time-tagged Cf source to compute the detector efficiency as a function of neutron energy.
Setting Bias Voltages and Thresholds

The goal for setting the bias voltage and thresholds for the $\alpha$ pixels associated with the DT generator is to maximize the true $\alpha$ count rate while minimizing the contribution of false $\alpha$ pulses. False $\alpha$ pulses are those that are not associated with a DT reaction and may come from noise from the PMT or background radiation. The threshold is determined by sending the signal from each $\alpha$ pixel to a spectroscopy amplifier and a multi-channel analyzer (MCA) that records the height of the pulses. Two spectra are recorded with the MCA. The first spectrum is ungated such that all the pulses from the $\alpha$ pixel are counted. For the second spectrum, a plastic radiation detector is placed in the path of the neutrons associated with the $\alpha$ pixel, and the MCA only counts the pulses from the $\alpha$ pixel that are in coincidence with a pulse from the plastic detector. The two spectra are shown in Figure 3-10. Notice that the pulses with higher energy are the ones associated with neutrons. The raw gated spectrum is smaller than the ungated spectrum because neutron detector and associated electronics are not 100% efficient. To allow a direct comparison to the ungated spectrum, a scaling factor is applied to the gated spectrum such that the two spectra agree well in the channels associated with neutrons. This scaled spectrum is shown in black in Figure 3-10. Using the ungated and scaled gated spectra, one can easily determine the threshold where the false $\alpha$ pulses only contribute $\sim 5\%$ to the total pulses recorded.

Of course, the spectra will shift to the right or left as the high voltage is changed. Experience with the particular DT generator and H9500 PMT used in the experiments has shown that a bias voltage of -1050V results in high count rates that are fairly evenly
divided among the pixels. For example, profiles shown in Figure 3-5 were taken with a bias voltage of -1050V, and each pixel has roughly the same count rate. The $\alpha$ pixel thresholds and pulse widths used in the experiments in this dissertation are shown in Table 3-2. The width of each pulse is not always exactly the same, so the data acquisition software accepts a range of widths set by the user.

The imaging detectors and the fission radiation detectors use the same process for setting the bias voltages and thresholds. A $^{137}$Cs source is used for the initial setup, and a $^{252}$Cf time-of-flight measurement is used for final fine-tuning.
Table 3-2. Thresholds and pulse widths for α pixels using bias voltage of -1050V

<table>
<thead>
<tr>
<th>Alpha Pixel</th>
<th>Threshold (mV)</th>
<th>Pulse Width Range (ns)</th>
</tr>
</thead>
<tbody>
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<td>dtp01</td>
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<td>52-68</td>
</tr>
<tr>
<td>dtp02</td>
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<td>72-88</td>
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<td>92-108</td>
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<td>dtp06</td>
<td>55.6</td>
<td>92-108</td>
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<td>dtp07</td>
<td>58.4</td>
<td>115-130</td>
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<td>dtp08</td>
<td>67.1</td>
<td>115-130</td>
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<td>dtp09</td>
<td>50.2</td>
<td>135-155</td>
</tr>
<tr>
<td>dtp10</td>
<td>53.5</td>
<td>135-155</td>
</tr>
<tr>
<td>dtp11</td>
<td>52.9</td>
<td>160-175</td>
</tr>
<tr>
<td>dtp12</td>
<td>52.4</td>
<td>160-175</td>
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<tr>
<td>dtp13</td>
<td>55.6</td>
<td>176-195</td>
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<tr>
<td>dtp14</td>
<td>61.8</td>
<td>176-195</td>
</tr>
<tr>
<td>dtp15</td>
<td>61.4</td>
<td>196-210</td>
</tr>
<tr>
<td>dtp16</td>
<td>45.5</td>
<td>196-210</td>
</tr>
</tbody>
</table>

Since eight image detectors share the same bias voltage, the detectors in the group have been gain matched using a $^{137}$Cs source. In other words, the detectors in the group have been chosen such that each gives a similar pulse height with the same voltage. The high voltage is adjusted so that the pulse height of the $^{137}$Cs Compton edge is between 120 and 150 mV for all the detectors in the group. Next, the threshold for each detector is set to $\sim 1/3$ of the pulse height of the $^{137}$Cs Compton edge. The goal for both the imaging and fission radiation detectors is to detect fast neutrons. However, the plastic detectors are also sensitive to gamma rays. Setting the threshold for each detector at $\sim 1/3$ of the pulse height of the $^{137}$Cs Compton edge prevents counting 186 keV gamma rays from unshielded uranium. In addition, this threshold also eliminates
counting any neutrons below 1 MeV since the light produced by a 159 keV gamma ray, which is approximately 1/3 the energy of the $^{137}$Cs Compton edge, is about the same as that from a 1 MeV neutron.

After the initial setup with the $^{137}$Cs source, the detectors are fine-tuned with a $^{252}$Cf time of flight measurement. The $^{252}$Cf source is enclosed in an ion chamber and is attached to a pre-amplifier. The ionization chamber uses +200 to 400V for bias voltage, and the amplifier requires +15V for power. Before being routed to the NMIS input boards as the synchronization signal, the signal out of the pre-amplifier passes through a CFD. $^{252}$Cf decays by $\alpha$ decay and spontaneous fission, and both decay modes are detected by the ion chamber. However, only the spontaneous fission events are of interest since $\alpha$ decays do not produce any neutrons. Since fission fragments produce larger pulses than $\alpha$ particles, the CFD threshold is adjusted to eliminate the counts due to $\alpha$ particles. An example of adjusting the threshold is shown in Figure 3-11. In this example, the signal from the CFD is input into a scalar, and the counts are measured with different threshold settings. As expected the count rate decreases as the threshold is increased. The steep drop in counts between 60 and 70 mV is due to removing the $\alpha$ decay contribution. The more gradual drop in counts after 80 mV is due to removing the contribution from fission fragments. In this particular example, the threshold was set at 85 mV.
For the imaging detectors, the $^{252}$Cf source is placed at the same position as the source of neutrons from the DT generator, 110 cm from each detector, so that each detector has the same solid angle. After making a short time correlation measurement, several tests are used to adjust the thresholds for the imaging detectors. First, the cross-correlation between the Cf source and each imaging detector is computed and plotted as shown in Figure 3-12. Recall that the plot shows the difference in time between when a Cf fission is detected and when a particle is detected in one of the scintillators. Since photons travel at $\sim30$ cm/ns, the gammas should arrive at the detectors at $\sim3.7$ ns after a fission event. Due to the time it takes for the signal to travel from the detector to being registered in the data acquisition boards, the gammas may not arrive at $\sim4$ ns.
So, if the gamma peak is not at ~4 ns for any of the detectors, the time skews in the detector configuration file need updating. In the second test, IDAS computes the neutron detection efficiency as a function of energy as shown in Figure 3-13. Notice that neutrons with energies less than 1 MeV are not counted. Mihalczo states that the detection efficiency of imaging detectors should be between 60% and 70% at ~2 MeV [30]. The peak detection efficiency for the plastic fission radiation detectors should also be around 65%. By examining these plots, the thresholds and bias voltages are adjusted to give each detector the desired efficiency and response. The bias voltages, thresholds, pulse widths, and pulse width ranges for all the plastic scintillator detectors used for the experiments in this dissertation are shown in Table 3-3 and Table 3-4.
Figure 3-13. Detector efficiency as a function of neutron energy for first eight imaging detectors
Table 3-3. Imaging detector settings

<table>
<thead>
<tr>
<th>Detector</th>
<th>Bias Voltage (V)</th>
<th>Threshold (mV)</th>
<th>Pulse Width Range (ns)</th>
</tr>
</thead>
<tbody>
<tr>
<td>id01</td>
<td>-903</td>
<td>40</td>
<td>45 - 65</td>
</tr>
<tr>
<td>id02</td>
<td>-903</td>
<td>29</td>
<td>45 - 65</td>
</tr>
<tr>
<td>id03</td>
<td>-903</td>
<td>37</td>
<td>45 - 65</td>
</tr>
<tr>
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<td>-903</td>
<td>41</td>
<td>45 - 65</td>
</tr>
<tr>
<td>id05</td>
<td>-903</td>
<td>45</td>
<td>45 - 65</td>
</tr>
<tr>
<td>id06</td>
<td>-903</td>
<td>39</td>
<td>45 - 65</td>
</tr>
<tr>
<td>id07</td>
<td>-903</td>
<td>58</td>
<td>74 - 95</td>
</tr>
<tr>
<td>id08</td>
<td>-903</td>
<td>60</td>
<td>74 - 95</td>
</tr>
<tr>
<td>id09</td>
<td>-856</td>
<td>36</td>
<td>74 - 95</td>
</tr>
<tr>
<td>id10</td>
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<td>40</td>
<td>74 - 95</td>
</tr>
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<td>41</td>
<td>74 - 95</td>
</tr>
<tr>
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<td>53</td>
<td>74 - 95</td>
</tr>
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<td>56</td>
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</tr>
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<td>100 - 131</td>
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<td>100 - 131</td>
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<td>38</td>
<td>100 - 131</td>
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<td>id19</td>
<td>-809</td>
<td>45</td>
<td>130 - 145</td>
</tr>
<tr>
<td>id20</td>
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<td>51</td>
<td>130 - 145</td>
</tr>
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<td>id21</td>
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<td>47</td>
<td>130 - 145</td>
</tr>
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<td>53</td>
<td>130 - 145</td>
</tr>
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<td>130 - 145</td>
</tr>
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<td>146</td>
<td>160 - 180</td>
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</table>
Table 3-4. Fission radiation detector settings

<table>
<thead>
<tr>
<th>Detector</th>
<th>Bias Voltage (V)</th>
<th>Threshold (mV)</th>
<th>Pulse Width Range (ns)</th>
</tr>
</thead>
<tbody>
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<td>-2200</td>
<td>44.2</td>
<td>24 - 40</td>
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</tbody>
</table>

Simulations with MCNP-PoliMi

The nuclear community uses many Monte-Carlo codes, but MCNP [33] and MCNPX [34] developed by Los Alamos National Laboratory are probably the most widely used codes. The primary difference between MCNP and MCNPX is that MCNPX can model charged particle transport. In earlier versions of MCNP and MCNPX, each neutron collision was not modeled with the best available physics information. First, MCNP and MCNPX did not sample the entire multiplicity distribution to determine the number of neutrons produced in a fission event. Instead, they only choose from the two integers nearest $\bar{\nu}$. For example, $\bar{\nu} = 2.42$ for induced fission of $^{235}$U by a thermal neutron. 58% of the time, the codes choose $\nu = 2$. 42% of the time, the codes choose $\nu = 3$. Secondly, after a neutron collision, MCNP and MCNPX sample secondary photon production before determining the type of the neutron collision. Therefore, the codes sometimes produce photons for neutron elastic scattering. While these two issues are trivial when calculating...
average values, such as fluxes, based on a large number of histories, they are important when trying to predict time correlation measurements between detectors. [35]

Even though later versions of MCNP and MCNPX corrected the sampling of the multiplicity distribution and secondary photon production, the ORNL group that operates NMIS already had significant experience using the MCNP-PoliMi code [35] when the research in this dissertation started. MCNP-PoliMi is built upon MCNP-4C code. Padavani and Pozzi corrected sampling of the multiplicity distribution and secondary photon production and tailored the code for time correlation measurements. They tried to implement physics that models each neutron reaction as close to reality as possible. Furthermore, they added distributions to the code that model spontaneous fission of $^{252}$Cf, $^{238}$U, $^{240}$Pu, $^{242}$Pu, $^{242}$Cm, and $^{244}$Cm.

In a simulation, each $\alpha$ pixel from the DT generator is modeled separately with its own input deck. The initial direction of the 14 MeV neutron in an input deck for a particular $\alpha$ pixel is chosen from a Gaussian fit to the data in Figure 3-5. So, a total of 16 MCNP-PoliMi runs (one for each $\alpha$ pixel) are necessary to simulate a measurement. Most MCNP-PoliMi runs in this dissertation include 10 million source particles and require between two and five hours of computational time on a 3.4 GHz Xeon processor.

For cells in the geometry that are identified as detectors, MCNP-PoliMi writes data for every collision in a special output file as shown in Table 3-5. For each collision, the file contains the history number, projectile type, collision type, target nucleus, energy deposition, time into the history, and location.
Table 3-5. Sample output From MCNP-PoliMi [35]

<table>
<thead>
<tr>
<th>History Number</th>
<th>Particle Number</th>
<th>Projectile type*</th>
<th>Interaction type†</th>
<th>Target nucleus</th>
<th>Cell number of collision event</th>
<th>Energy deposited in collision (MeV)</th>
<th>Time (shakes)</th>
<th>Collision position</th>
<th>Particle weight</th>
<th>Generation number</th>
<th>Number scatterings</th>
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<td>1001</td>
<td>4</td>
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<td>0.65</td>
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<td>4.8</td>
<td>11.02</td>
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<td>0.748</td>
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<td>8.49</td>
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<td>5</td>
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<td>1</td>
<td>-99</td>
<td>1001</td>
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<td>0.0279</td>
<td>4.604</td>
<td>21.85</td>
<td>-6.07</td>
<td>1.26</td>
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<td>5</td>
<td>0.00551</td>
<td>5.465</td>
<td>21.87</td>
<td>-6.73</td>
<td>0.53</td>
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<td>1</td>
</tr>
<tr>
<td>910</td>
<td>1</td>
<td>1</td>
<td>-99</td>
<td>1001</td>
<td>5</td>
<td>5.75385</td>
<td>0.656</td>
<td>17.03</td>
<td>-0.65</td>
<td>3.84</td>
<td>1</td>
<td>0</td>
</tr>
</tbody>
</table>

* 1 = neutron; 2 = photon
† Δ -99 = elastic scattering; 1 = Compton scattering
● 1001 and 1 = hydrogen; 6000 = carbon
In his dissertation, Grogan provides a FORTRAN program, PoliMiPP, that computes the singles and doubles distributions from the data in the MCNP-PoliMi output file [28]. A newer version of PoliMiPP is used for the post processing in this dissertation and is attached. The program mimics the way an actual detector responds and allows the user to specify a pulse generation time, a dead time, and a light threshold. For each history, the program determines which detector cells had a collision and the times between the collisions. If two collisions occur within the pulse generation time setting, the energy deposited is combined into one pulse. Grogan recommends setting the pulse generation time to twice the rise time of the pulses coming from a particular detector. After combining appropriate collisions, the program determines if the pulse occurred during dead time. If the pulse occurs within the dead time after a previous pulse, the pulse is not counted. Finally, the program determines if the height of the pulse would pass the threshold of the CFD. PoliMiPP allows the user to specify a neutron threshold in MeV and uses an equation from Pozzi, Mullens, and Mihalczo [36] to convert the neutron threshold into a light threshold. The equation is

\[ L = A_0 + A_1 E_n + A_2 E_n^2, \]  

where \( L \) is the light in MeVee and \( E_n \) is the neutron energy in MeV. The values of the coefficients are \( A_0 = 0, A_1 = 0.125, \) and \( A_2 = 0.0364. \) The unit MeVee or MeV electron equivalent is used to reflect the fact that different particles with the same energy deposition in a scintillator will produce different amounts of light [37]. By definition, the light produced by an electron with an energy deposition of 1 MeV is 1MeVee. Equation 3-2 is also used to convert energy deposited by neutrons into light. For gamma
rays, Pozzi, Mullens, and Mihalczko show that the light generated by photons in MeVee is approximately the same as the energy deposited [36]. In PoliMiPP, 1 MeV = 1 MeVee for gamma rays. If the collision generated enough light to pass the threshold, PoliMiPP records a detection.

PoliMiPP has some features to account for statistical variations in the pulse height and the threshold. The pulse height has statistical fluctuations because there are random fluctuations in the number of photons produced in the detector and the number of photoelectrons produced in the PMT. The threshold also has some small statistical fluctuations. For example, if 100 identical pulses having a height equal to the CFD threshold were passed to the CFD, not all of the pulses trigger the CFD. To account for these statistical variations, PoliMiPP samples Gaussian distributions for each pulse to determine a particular pulse height and threshold.

PoliMiPP first accounts for the statistical variation in the pulse height. After determining all the deposited energy in a pulse and converting it to MeVee, PoliMiPP calculates the number of photoelectrons produced in the pulse. PoliMiPP allows the user to input the average number of photoelectrons produced per MeVee of deposited energy. This parameter is named $P_{E per MeV}$. For plastic scintillators, Grogan recommends a value of 900 photoelectrons per MeVee. So, the photoelectrons produced in a pulse are

$$Num_{PE} = P_{E per MeV} \cdot PH,$$

where $Num_{PE}$ is the number of photoelectrons produced and $PH$ is the energy deposited in the pulse in units of MeVee. To account for the statistical variation of the
number of photoelectrons produced, PoliMiPP samples $\Delta PE$ from a standard normal distribution whose standard deviation is $\sqrt{NumPE}$. Therefore, the sampled number of photoelectrons in the pulse is

$$NumPE_{sample} = NumPE + \Delta PE.$$  \hspace{1cm} 3-4

Finally, the sample number of photoelectrons is converted back to a pulse height in units of MeVee by

$$PH_{sample} = \frac{NumPE_{sample}}{PEperMeV}. \hspace{1cm} 3-5$$

After accounting for the statistical variation in the pulse height, PoliMiPP accounts for the statistical variation in the threshold. Again, PoliMiPP takes an input parameter, $SDTHRESH$, which is the standard deviation of the threshold in the units of MeV. The neutron threshold and $SDTHRESH$ are determined by fitting a simulation to a $^{252}$Cf time of flight measurement. As before, PoliMiPP samples $\Delta Thresh$ from a standard normal distribution whose standard deviation is $SDTHRESH$. So, the sampled threshold is

$$Thresh_{sample} = Thresh + \Delta Thresh,$$ \hspace{1cm} 3-6

where $Thresh$ is the neutron threshold in units of MeV. This sampled threshold is converted to MeVee using Equation 3-2, and if $PH_{sample} \geq Thresh_{sample}$, then the pulse is recorded as a count.

After all histories have been processed, PoliMiPP writes the resulting distributions to output files. Since each $\alpha$ pixel has its own input deck and PoliMiPP output files, the singles and doubles distributions in the output files are summed before comparison to a measurement.
CHAPTER 4

FEASIBILITY STUDIES

Feasibility studies were performed before any actual measurements. The feasibility studies primarily show how the time correlation distributions differ for different materials and geometrical configurations. Note that the feasibility studies do not include the actual as-built dimensions of the depleted uranium shields nor the exact geometrical arrangement as the actual measurements. Furthermore, the fission radiation detectors are modeled as liquid instead of plastic scintillation detectors. Early in the research, liquid scintillators were planned for the experiments to take advantage of pulse shape discrimination. With pulse shape discrimination, the detector pulses from neutrons can be separated from pulses due to gamma rays. As a result, the studies in this chapter only include the contribution from neutrons. Due to a desire for near real-time pulse shape discrimination and difficulty with the NIM modules that perform the pulse shape discrimination, use of the liquid detectors was abandoned. Since the gamma rays and neutrons are predominately observed at different times following interrogation, the pulse shape discrimination was not necessary for these measurements.

Baseline Configuration

In the baseline configuration, the inner uranium casting is modeled with enrichments of 93%, 70%, 50%, 30%, and 0.3%. Each shield is depleted uranium with a thickness of 0.5 in. and a height equal to the inner uranium casting. There is a 1/8 in. gap between the inner uranium casting and the first shield as well as between the remaining
shields. The dimensions of each shield are given in Table 4-1, and a plan view of the casting and the shields is shown Figure 4-1. The inner uranium casting and shields are centered inside a 55 gallon drum, and the remaining space inside the drum is filled with Celotex®. Celotex® is a material that is sometimes used at Y-12 National Security Complex to protect items in shipping containers [38].

<table>
<thead>
<tr>
<th></th>
<th>Inner Shield</th>
<th>Middle Shield</th>
<th>Outer Shield</th>
</tr>
</thead>
<tbody>
<tr>
<td>Height (in)</td>
<td>6</td>
<td>6</td>
<td>6</td>
</tr>
<tr>
<td>Inside Radius (in)</td>
<td>2.625</td>
<td>3.25</td>
<td>3.875</td>
</tr>
<tr>
<td>Outside Radius (in)</td>
<td>3.125</td>
<td>3.75</td>
<td>4.375</td>
</tr>
<tr>
<td>Thickness (in)</td>
<td>0.5</td>
<td>0.5</td>
<td>0.5</td>
</tr>
</tbody>
</table>

Figure 4-1. Plan view of inner uranium casting surrounded by layers of shielding
The geometrical arrangement of the DT generator, the imaging detectors, and the fission radiation detectors with respect to the inner uranium casting and shields is shown in Figure 4-2. The DT generator is modeled as a point source located 35 cm from the center of the inner uranium casting and at the midheight of the casting. The fission radiation detectors are in a 2x2 arrangement as shown in Figure 4-2 with each arrangement 68 cm from the center of the inner uranium casting. There are 6 inches of polyethylene between the detectors in each arrangement. The polyethylene is present to reduce the chance that a neutron collides in one detector and enters another.

Figure 4-3 shows the singles distribution for one DU shield and several enrichments. For clarity, the contribution from any gamma rays has been removed from the distribution and all following distributions in this chapter unless otherwise noted. The peak at approximately 22 ns is due to uncollided neutrons from the DT generator. The decline from approximately 22 to 30 ns is primarily due to scattered neutrons arriving at the detectors. Each enrichment has about the same amount of uncollided and scattered neutrons arriving at the detectors. Fission neutrons start arriving at the detectors at approximately 30 ns. Since the fission cross section for $^{235}$U is greater than $^{238}$U, as shown in Figure 2-1, higher enrichments produce more fission neutrons. Each simulated enrichment in Figure 4-3 is easily distinguishable from the others, especially considering that the vertical axis is on a logarithmic scale. So, if a measured distribution aligned closely with one of the lines in the plot, the line would give an estimate of the actual enrichment.
Figure 4-2. Arrangement of the object, fission radiation detectors, and DT generator in feasibility studies. The drum and Celotex® are not shown.
The doubles distribution is shown in Figure 4-4. Since the sole interest is fission neutrons, only neutrons that arrive at the detectors at times between 35 and 70 ns are counted in the doubles distribution. Once again, notice that each enrichment has a distinct signature.
Figure 4-4. Doubles distribution for one DU shield in baseline configuration

The final time signature of interest combines information from the singles and the doubles distributions and is called the Feynman distribution in this dissertation since it is related to the Feynman variance [39]. The distribution is the integral of the doubles distribution from time 0 to $t$ ns divided by the total number of fission neutrons from the singles distribution. Mathematically, the distribution is calculated by

$$y(t) = \frac{\sum_{t'=0}^{t} Doubles(t')}{\sum_{t'=35}^{70} Singles(t')}$$

4-1
where $y(t)$ is the Feynman distribution, *Doubles* is the doubles distribution, and *Singles* is the singles distribution. As before with the doubles distribution, the summation limits in the denominator are chosen to remove any contribution from uncollided and scattered neutrons. Croft *et al.* provide an excellent derivation showing how the ratio of the doubles to the singles is related to the Feynman variance [40]. The Feynman distribution for one DU shield is shown in Figure 4-5.

The plots in Figure 4-6 show the effect of adding additional shielding on the time signatures. There are many lines in the plots that make them somewhat difficult to read. However, the purpose of the plots is to demonstrate the possibility of overlapping distributions if the geometry is unknown. If the geometry is known, then the irrelevant cases would be removed and the plots would appear as in Figure 4-3 through Figure 4-5. Nevertheless, notice that some cases in the doubles distribution are very close to each other. For example, the 93% enriched case with three shields is similar to the 70% enriched case with only the inner shield present. However, in the Feynman distribution, the cases are far apart. In fact, the Feynman distribution in Figure 4-6 seems to suggest that each enrichment case approaches an asymptotic value regardless of the shielding layers present. If true, then exact knowledge of the geometrical arrangement from the neutron image may not be necessary.
Figure 4-5. Feynman distribution for one shield in baseline configuration.

The x-axis shows the time difference between the detection of the first and second neutron.
Figure 4-6. Time signature distributions for the baseline configuration
The line type indicates the shielding layers present (dots, inner only; dashes, inner and middle; and solid, all three layers).
Different Shielding Materials

To understand how different materials affect the time signatures, the shields were modeled as lead and steel in simulations and compared to the baseline configuration with DU shields. The geometry remained the same as the baseline configuration. The resulting time signatures are shown in Figure 4-7 through Figure 4-9. In the plots, the color of the lines indicates the enrichment of the inner HEU casting, and the line type indicates the shielding material. Solid lines represent the baseline DU shields, dashed lines represent lead shields, and dotted lines represent steel shields. The top plot in each figure shows the distributions for 0.5 inches of shielding, and the bottom plot shows the distributions for 1.5 inches of shielding. Again, there are many lines on the plots that make them somewhat difficult to read. However, the goal for examining the different cases of shielding material and thicknesses is to determine if each case is sufficiently unique to avoid mistaking it for another case. If each case is unique, then it may be possible to identify the enrichment without any prior knowledge of the geometry and materials present.

The singles distributions are shown in Figure 4-7. While none of the cases are exactly the same, several are very similar. For example, in the top plot, the distribution for the 93% enriched casting with steel shields is very close to the one for the 70% enriched casting surrounded by DU. These two cases are highlighted in Figure 4-7 with a blue arrow. Similarly, the distribution for the 70% enriched casting with steel shields is very close to the one for the 50% enriched casting surrounded by DU. Of course, if the
Figure 4-7. Singles distributions for different shielding materials and different thicknesses. The line type indicates the shield material (solid, depleted uranium; dashes, lead; and dots, steel).
Figure 4-8. Doubles distributions for different shielding materials and different thicknesses
The line type indicates the shield material (solid, depleted uranium; dashes, lead; and dots, steel).
Figure 4-9. Feynman distributions for different shielding materials and different thicknesses. The line type indicates the shield material (solid, depleted uranium; dashes, lead; and dots, steel).
tomographic images allow identifying the shielding material, then comparing the simulated time correlations with different shielding materials is unnecessary.

The doubles distributions are shown in Figure 4-8, and as with the singles distributions, many overlap. In the top plot, the distributions for the 70% enriched casting surrounded by steel shields and the 50% enriched casting surrounded by depleted uranium still are very close. The 93% enriched cases are distinct in the top plot but not in the bottom plot. In the top and bottom plots, the 70% enriched casting with steel shields is the same as the 30% casting with DU shields.

The Feynman distributions are shown in Figure 4-9. Recall that the Feynman distribution combines information from both the singles and doubles distribution and may spread out overlapping cases in those distributions. Indeed, none of the cases in top plot overlap although resolving the difference between the 50% enriched casting with steel shielding and the 30% enriched casting with depleted uranium may be difficult. There are about two more overlapping cases in the bottom plot.

While some cases may overlap in one or two of the distributions, none overlap in all three. However, there are an infinite number of combinations of different shielding thicknesses and materials, and it is possible that some will overlap in all three distributions. Even with the cases presented in this study, it is difficult to examine all three distributions to find features that uniquely distinguish each case. If a neutron image showed that the shielding thickness is 1.5 inches, then there would be no need to examine the top plot. Secondly, if the attenuation coefficients from the neutron image showed that the shielding material was steel, then the dashed and solid lines in the
bottom plots could be removed. Then, only five cases where the enrichment changes would remain in each plot. Comparing each of the remaining five cases with a measurement is much simpler than having to consider all of the cases in this study.

**Irresolvable Gap between the Casting and DU Shield**

Since this method of determining enrichment is dependent on the neutron image, the method is sensitive to how well the geometry can be inferred from the image. In particular, if the resolution is not sufficient to identify the gap separating the inner casting from the shield and the shield is depleted uranium, the casting and shield will appear as one object since enriched uranium and depleted uranium have the same 14 MeV attenuation coefficient. Figure 4-10 shows the Feynman distribution where the casting and shield are modeled as one object. In all cases, the thickness of the combined object is 2 in., but the thickness of the enriched part is modeled as ½, ⅔, and 1 in. thick. Notice that ¾ in. of 93% HEU has the same distribution as 1 in. of 70% HEU. These cases also overlap in the singles and doubles distributions. Therefore, the boundary separating the enriched uranium from depleted uranium must be identifiable.
HEU on Outside of DU

Another possible combination of the enriched casting and shield puts the casting outside the shield. Of course, this should not happen in reality, but one purpose of verification measurements is to identify any abnormalities. In this situation, gamma spectroscopy may also be able to identify the enrichment. Nevertheless, Figure 4-11 shows the Feynman distribution for a $\frac{3}{8}$ in. thick inner casting and a $\frac{1}{2}$ in. thick outer casting. The solid lines represent the cases where the inner casting is enriched uranium and the outer casting is depleted uranium. Conversely, the dotted lines represent the cases where the inner casting is depleted uranium and the outer casting is enriched.
uranium. The case with 70% enriched uranium on the inside is indistinguishable from the case of 93% enriched uranium on the outside. Since enriched uranium appears the same as depleted uranium in the neutron image, fission mapping as described by Hausladen [41] is needed to distinguish the time signatures. Fission mapping shows the location and intensity of fissions throughout an object and therefore should be able to show the location of the enriched uranium.
Increasing Thickness of Enriched Casting

In the next set of simulations, the thickness of the inner casting is varied while holding the enrichment and shield thickness constant. The inside radius of the casting is constant at 1.75 in., and the thickness is increased from the baseline thickness of 0.75 in. to 2.00 in. with 0.25 in. steps. To allow for the increasing thickness of the inner casting, only the outer depleted uranium shield is in the model. The enrichment of the inner casting is 30%. The Feynman distributions for this set of simulations are shown in Figure 4-12. As the thickness of the casting increases, more neutrons cause fission, and the Feynman distribution increases.

Figure 4-12. Feynman distribution for varying inner casting thickness and constant shield thickness
The enrichment of inner casting is constant at 30%. The thickness of the DU shield is constant at 0.5 in.
**Increasing Thickness of Outer Shield**

In this set of simulations, the thickness of the outer depleted uranium shield is increased from the baseline configuration of 0.5 in. to 4 in. thick in 0.5 in. increments. The other two shields are still present in the model, and the enrichment of the inner casting is 93%. Figure 4-13 shows the resulting Feynman distributions. As the outer shield thickness increases, fewer neutrons are able to reach the inner enriched casting. In addition, fewer fission neutrons are able to escape the shielding and reach the detectors. Therefore, the Feynman distribution decreases as the thickness increases.

![Feynman distribution for increasing thickness of outer DU shield](image-url)
Plastic Detectors and New Detector Arrangement

As discussed in the introduction to this chapter, the liquid scintillator detectors were abandoned for plastic detectors. In addition, the arrangement of the fission radiation detectors was changed to that shown in Figure 4-14. The new arrangement increases the solid angle of the detectors and allows them to detect more fission neutrons. The fission radiation detectors are aligned on an arc that is approximately 48 cm from the centerline of the stand. As with the 2x2 arrangement, polyethylene blocks are positioned between the detectors to minimize cross talk. Figure 4-15 compares the Feynman distribution for the baseline configuration in the 2x2 arrangement with liquid detectors to the new arrangement with plastic detectors. The shape of the distributions in both arrangements is the same. However, the vertical limit for the new arrangement is almost twice that for the old. Nevertheless, Figure 4-15 shows that the studies using the liquid detectors have merit even though the plastic detectors are used in the experiments.
Figure 4-14. New arrangement of fission radiation detectors
Figure 4-15. Comparison of new to old detector arrangement
The line type indicates the shielding layers present (dots, inner only; dashes, inner and middle; and solid, all three layers).
Summary

The studies presented in this chapter indicate that the proposed method of determining enrichment is indeed feasible. If the materials present and geometry are known from the tomographic images, each simulated enrichment gives a unique time correlation signature. Without the material and geometrical information from the tomographic images, the signatures are not necessarily unique. Therefore, determining the enrichment is more tractable with information from the images.
CHAPTER 5
MEASUREMENTS AND SIMULATIONS

This chapter discusses the experiments used to test the proposed method to determine uranium enrichment and compares the experimental results to simulations. In the primary measurements, neutrons from the DT generator interrogate the castings and shields. Tomographic images based on the response recorded by the imaging detectors and the large fission radiation detectors are used to extract the geometrical and material estimates of the castings and shields. In the next set of measurements, neutrons from a $^{252}\text{Cf}$ source interrogate the HEU casting and shields, and the response is recorded by small 1 x 1 x 6 in. plastic scintillator detectors. The geometrical and material estimates from the primary measurements are applied to simulations of the latter measurements and are used to predict the enrichment of the HEU casting.

The primary casting measurements were performed at Site 1 of the Y-12 Nuclear Detection and Sensor Test Center [42]. A DU casting was measured on February 23, 2012, and a 93.186 weight percent $^{235}\text{U}$ casting was measured on February 27, 2012. Both annular castings have a ~6 in. height, 3.50 in. inner diameter, and 0.750 in. thickness. The HEU casting has a mass of 17.920 kg, and the DU casting has a mass of 17.146 kg. Both are canned inside 0.025 in. thick stainless steel to prevent contamination. The HEU casting was in an annular can, whereas the DU casting was in a cylindrical can.

Each casting was measured bare and with up to three layers of DU annular shields. The shields are shown in Figure 5-1, and their dimensions are given in Table 5-1. The
The arrangement of the detector system used in the primary measurements is shown in Figure 5-2. The height of the cart holding the casting and DU shields was adjusted to position the mid-height of the casting at 70.5 cm above the floor. This height puts the casting in the middle of the two rows of fission radiation detectors. The horizontal center of the casting was 27.8 cm from the center of the target in the neutron generator along a line in the direction of imaging detector position 72 (see Figure 3-5 for...
the imaging detector position). The stand holding fission radiation detectors was centered on the castings and rotated such that the distance from each corner of the stand to the center of the target within the neutron generator was the same.

Each casting and shield combination was measured for approximately 30 minutes. After the bare casting was measured, each layer of shield was subsequently added for a measurement.

**Time Correlations from Fission Radiation Detectors**

The time distribution of single events in any fission radiation detector following the detection of an α particle from the neutron generator is shown in Figure 5-3 for the bare castings. The first peak at ~4 ns is due to gammas from inelastic scattering of the 14 MeV neutrons within the neutron generator. Since this peak is related solely to the number of 14 MeV neutrons produced, it has the same height for both measurements.
Next, the 14 MeV neutrons reach the casting and some cause fission. The second peak, which is about 9 ns, is due to fission prompt gammas reaching the detectors. Since this peak is associated with fission in the casting, the peak for the HEU casting is higher than the one for the DU casting. At ~18 ns, the 14 MeV neutrons that did not interact with the castings reach the detectors. Again, the peak in both measurements has the same height. Shortly after 18 ns, scattered neutrons arrive at the detectors. Finally, particles detected after about 30 ns are primarily fission neutrons.

The time distribution of singles events for all the casting and shield combinations measured are shown Figure 5-4. The measurements with the HEU casting are shown in red, and the measurements with the DU casting are shown in blue. The line type on the
Figure 5-4. Distribution of singles after source event for all combinations of castings and shields (The plot includes the contribution from all pixels in the generator as well as all fission radiation detectors. In the legend, the letter “I” indicates the inner DU shield is present, the letter “M” indicates the middle shield is present, and the letter “O” indicates the outer shield is present.)

plot indicates the shields present. In the legend, the letter “I” indicates the inner DU shield is present, the letter “M” indicates the middle shield is present, and the letter “O” indicates the outer shield is present. Notice that more fission gammas reach the detectors if no shielding is present. Regarding fission neutrons, the overall trend is that more fission neutrons are detected as shielding is added because of increased multiplication and increased amount of fissionable material. As expected, the HEU casting produces more fission gammas and neutrons than the DU casting.

The time distributions for doubles events for all the casting and shield combinations measured are shown in Figure 5-5. This plot shows the time between two detections in the top row fission radiation detectors 32 to 100 ns after the alpha detection. For the HEU, there are more doubles detected with just the bare casting.
Since most fission occurs in the casting, adding layers of shielding decreases the chances that at least two fission neutrons from the casting will reach the detectors. All of the cases with the DU casting have approximately the same doubles distribution.

**Tomographic Reconstructions**

The tomographic reconstructions play an important role in estimating the enrichment since they permit the essential estimates of the materials and geometry present in a measurement. Figure 5-6 through Figure 5-9 show transmission images created using the filtered backprojection technique for the cases with the HEU casting. The transmission images are created by comparing the number of uncollided 14 MeV neutrons that reach the imaging detectors with material present to the number in a void measurement. These images show the neutron attenuation coefficient throughout a slice...
Figure 5-6. Filtered backprojection image of HEU casting without any shields

Figure 5-7. Filtered backprojection image of HEU casting with inner shield
Figure 5-8. Filtered backprojection image of HEU casting with inner and middle shields

Figure 5-9. Filtered backprojection image of HEU casting with all shields
at the midheight of the casting and shields. Since HEU and DU have essentially the same attenuation cross section, the HEU parts cannot be distinguished from the DU parts in Figure 5-7 through Figure 5-9.

To extract the geometry and attenuation coefficients from the transmission images, a computer code called Fitting is used [43]. This code applies pattern recognition algorithms to determine the appropriate boundaries for each region and the best attenuation coefficient for each region. Table 5-2 shows the estimates from Fitting for the outer diameters in each region of the tomographic images as well as the true dimensions. Overall, the algorithm in Fitting estimates the geometry very well. The mean error from the true dimensions is 0.26 cm, and the maximum error is 0.59 cm. The estimates of the attenuation coefficients in each region are given in Table 5-3. Again, the algorithm did well. The 14 MeV attenuation coefficient for both HEU and DU is 0.28 cm$^{-1}$, so the estimates for the uranium regions are a little low. Similarly, the estimates in some of the void regions are a little high. The worst estimate, 0.16 cm$^{-1}$, is for the void region between the casting and inner shield for the measurement where both the inner and middle shields were present. It is possible that this region could be interpreted to contain a material other than air.

Even though HEU cannot be distinguished from DU in transmission images, Hausladen describes a reconstruction technique that shows the fission density throughout the object [41]. This technique involves creating projections based on the number of doubles detected from each 14 MeV interrogating neutron. The number of doubles detected from fission in a particular volume element is dependent
upon the attenuation of the 14 MeV neutron to the volume element, the fission probability of the volume element, the attenuation of any induced fission neutrons from the volume element to the detectors, and solid angle of the detectors. Hausladen shows this is a non-linear problem. Therefore, an iterative reconstruction method, such as the maximum likelihood expectation maximization method, must be used with the technique.

The results of Hausladen’s fission mapping technique are shown in Figure 5-10 through Figure 5-13 for each casting and shield combination. In each figure, the images on the top row are from measurements with the HEU casting, and the images on the
Figure 5-10. Reconstructions for castings without any shields
Left column shows transmission images, middle column shows doubles images, and the right column overlays the doubles image on the transmission image.

Figure 5-11. Reconstructions for castings with inner DU shield
Left column shows transmission images, middle column shows doubles images, and the right column overlays the doubles image on the transmission image.
Figure 5-12. Reconstructions for castings with inner and middle DU shields
Left column shows transmission images, middle column shows doubles images, and the right column overlays the doubles image on the transmission image.

Figure 5-13. Reconstructions for castings with all DU shields
Left column shows transmission images, middle column shows doubles images, and the right column overlays the doubles image on the transmission image. The gaps between the layers are clearly visible.
bottom row are from measurements with the DU casting. The images on the left side are the transmission images plotted with a grey scale. The middle images in the figures are reconstructions based on the number of correlated double detections in the fission radiation detectors in the time window of 35 to 80 ns after the creation of each 14 MeV neutron. These images show probability per unit length of creating two neutrons that are detected for each pixel in the image. Therefore, they essentially show the locations where fission occurs. In each figure, the color intensity is scaled based upon the maximum probability per length in the reconstruction with the HEU casting. The images on the right side show the doubles images (the middle images) overlaid on the transmission images. The overlaid images allow one to easily determine which component is causing the fission.

In the feasibility studies, there was a concern that the gap between each layer of uranium may not be visible in the transmission images. As the transmission images show, the gap indeed is clearly visible. In addition, there was a concern that the HEU component may not be located on the inside of the assembly, and if true, fission mapping may be needed to determine the location of the HEU. The HEU casting is clearly visible in overlaid images.

**Small Detectors and HEU Casting Interrogated with $^{252}$Cf**

The second set of measurements was made at the Y-12 Nuclear Detection and Sensor Test Center in July 2012. At this time, operations involving the neutron generator were not permitted at the test center, so neutrons from a $^{252}$Cf source were used to
interrogate the HEU casting and shields. Additionally, these measurements used four small 1x1x6 in. plastic detectors inside 0.25 in. thick lead sheaths to measure the fission radiation. In the past, simulated and measured time correlations involving interrogation with $^{252}\text{Cf}$ and small detectors have agreed well [44].

To determine the neutron threshold that should be used with the post processor, the small detectors were placed on top of a cart whose height was $\sim 102$ cm above the floor. The detectors were arranged in an arc such that the distance to the $^{252}\text{Cf}$ source was 112.5 cm for each detector. The measurement lasted $\sim 20$ minutes, and the simulation included 40 million spontaneous fissions. The simulated and measured time correlations are compared in Figure 5-14, and they agree very well. Notice that even the room return is accurately predicted in the simulation. The simulations were made with the code MCNPX-PoliMi [45], and the neutron threshold used with the post-processor is 1.2 MeV.

Using the setup of past measurements as a guide, the four small detectors were placed on the outside of the casting and shield assembly as shown in Figure 5-15. The height of the top of the cart supporting the assembly and detectors was $\sim 103$ cm above the floor, and the $^{252}\text{Cf}$ source was centered inside the HEU casting at a height of approximately 3¼ in. above the cart top. The singles distribution for the case with the HEU casting surrounded by all three DU shields is shown in Figure 5-16 with the color red being the measurement and black being the simulation. The simulation has excellent agreement with the measurement. Since the canning of the DU casting at Y-12 prevents placing the $^{252}\text{Cf}$ source in the middle of the casting, the measurements could not be
Figure 5-14. $^{252}$Cf time of flight measured by small fission radiation detectors

Figure 5-15. Arrangement for the measurements of the HEU casting and DU shields using $^{252}$Cf and small detectors
Figure 5-16. Results for $^{252}$Cf using all three DU shields and small detectors

repeated with the DU casting. However, a measurement was made with all three DU shields and no casting present. The measured and simulated distributions for this case are also shown in Figure 5-16 with the colors purple and blue, respectively. Again, there is excellent agreement. Finally, Figure 5-17 through Figure 5-19 show the excellent agreement between the measured and simulated singles distributions for the other shielding combinations of the HEU casting.

The doubles distributions for the simulations and measurements with the small detectors are shown in Figure 5-20 for all cases considered. In the plot, the measurements are indicated with dots and the simulations with solid lines. The plot includes the doubles detected between 2 and 50 ns after the $^{252}$Cf spontaneous fission. As shown, the simulations agree perfectly with the measurements.
Figure 5-17. Results for $^{252}$Cf interrogation of bare HEU casting using small detectors

Figure 5-18. Results for $^{252}$Cf interrogation of HEU casting and inner shield using small detectors
Figure 5-19. Results for $^{252}$Cf interrogation of HEU casting and inner and middle shields using small detectors.

Figure 5-20. Doubles distributions for measurements with small detectors. The legend indicates the casting present followed by the shielding layers present.
The simulations shown in Figure 5-16 through Figure 5-20 all use the actual dimensions of the casting and shields. With the correct dimensions and enrichments, the data show that simulations can accurately predict the time response of small detectors to fission neutrons induced by $^{252}$Cf neutrons.

**Predicting Enrichment of HEU Casting**

To demonstrate the proposed method of determining the enrichment of shielded uranium, the dimensions of the casting and shields from the Fitting estimates in Table 5-2 are used in simulations that predict the time correlations expected from the small detectors when the casting is interrogated by neutrons from a $^{252}$Cf source. The casting is modeled with enrichments of DU, 20%, 40%, 60%, 90%, and 95%, and the shields are modeled as DU. Other than changing the dimensions of the casting and shields to match those given in Table 5-2 and varying the enrichment of the casting, the simulations are the same those used presented in the previous section.

The first enrichment estimate is for the casting with no shields. The comparison of the measured singles distribution to the simulated distributions is shown in Figure 5-21. In the figure, the measured data is shown with red dots and each solid line represents a simulation with an enrichment noted in the legend. The measured singles distribution aligns well with the simulations where the enrichment is greater than 80%. It is clear that the enrichment is not less than 80%. The comparison of the measured doubles distribution to the simulated distributions is shown in Figure 5-22. Similar to the singles distribution, the measured doubles distribution aligns well with the prediction for the
Figure 5-21. Comparison of the measured singles distribution to simulated distributions with differing enrichments for the bare casting

Figure 5-22. Comparison of the measured doubles distribution to simulated distributions with differing enrichments for the bare casting

97
80% enriched casting. In the region between -3 and 3 ns, the measured values are closer to the simulation with the 90% enriched casting. To aid in estimating the enrichment, the total doubles measured is compared to the total doubles predicted in the simulations in Figure 5-23. The total doubles from the measurement is shown with a dashed red line spanning all enrichments. Error bars representing the measured doubles ± one standard deviation are too small to be seen in the plot. The total doubles from the simulations are shown in blue and include ± one standard deviation error bars. The red line from the measurement intersects with the blue line from the simulations at an enrichment of 82.5%. Considering the error bars, the estimate is 82.5% ± 0.4%. Even though this estimate differs from the actual enrichment by approximately 11%, the accuracy of the estimate is likely acceptable for many nonproliferation applications, including arms control and treaty verification activities where the goal may be simply to identify the presence of HEU.

The next enrichment estimate is for the case with the casting and inner shield. The comparison of the singles distributions is shown in Figure 5-24, and the comparison of the doubles distributions is shown in Figure 5-25. In the figures, the measured data align very well with the simulations where the casting is modeled as 90% and 95% enriched. The comparison of the total doubles is shown in Figure 5-26. The line from representing the measured total doubles intersects with the line from the simulations at an enrichment of 92.5%. Considering the error bars, the estimate is 92.5% ± 0.7%, which is very close to the actual enrichment of the casting.
Figure 5-23. Comparison of the total doubles from simulations with varying enrichments to the measured value for the bare casting.

Figure 5-24. Comparison of the measured singles distribution to simulated distributions with differing enrichments for the case of the casting and inner shield.
Figure 5-25. Comparison of the measured doubles distribution to simulated distributions with differing enrichments for the case of the casting and inner shield

Figure 5-26. Comparison of the total doubles from simulations with varying enrichments to the measured value for the case of the casting and inner shield
The third enrichment estimate is for the case with the casting and inner and middle shields. The singles and doubles distributions for this case are shown in Figure 5-27 and Figure 5-28, respectively. Once again, the measured data align very well with the simulations where the casting is modeled as 90% and 95% enriched. The comparison of the total doubles is shown in Figure 5-29. Based on the total doubles, the estimated enrichment of the casting is $94.5\% \pm 1\%$.

**Figure 5-27.** Comparison of the measured singles distribution to simulated distributions with differing enrichments for the case of the casting and inner and middle shields.
Figure 5-28. Comparison of the measured doubles distribution to simulated distributions with differing enrichments for the case of the casting and inner and middle shields.

Figure 5-29. Comparison of the total doubles from simulations with varying enrichments to the measured value for the case of the casting and inner and middle shields.
The final enrichment estimate is for the case with the casting and all shields. The singles and doubles distributions for this case are shown in Figure 5-30 and Figure 5-31, respectively. In the singles distribution, the measured data are slightly above the simulated data for the 95% enriched casting. In the doubles distribution, the measured data are predominately between simulated data for the 80% and 90% enriched casting. The comparison of the total doubles is shown in Figure 5-32. Based on the total doubles, the estimated enrichment of the casting is 88% ± 1%.

Figure 5-30. Comparison of the measured singles distribution to simulated distributions with differing enrichments for the case of the casting and all shields.
Figure 5-31. Comparison of the measured doubles distribution to simulated distributions with differing enrichments for the case of the casting and all shields.

Figure 5-32. Comparison of the total doubles from simulations with varying enrichments to the measured value for the case of the casting and all shields.
For the proposed method of determining the enrichment to work, the simulations must be able to accurately predict the measured distributions. With the small detectors, the simulations are indeed accurate. Appendix A compares simulations to data from measurements in February 2012 with the large fission radiation detectors, and the simulations do not accurately predict the measurements. Some attempts to improve the agreement are discussed in Appendix B, but further research is needed to improve the modeling of the large detectors.
CHAPTER 6

CONCLUSIONS AND FUTURE WORK

This dissertation presents a novel approach to determine the enrichment of uranium metal when shielded by high-Z materials. Other approaches typically rely on the detection of low-energy photons from uranium or uranium daughters and fail when the uranium is shielded. In the proposed method, time and directionally tagged neutrons from a DT generator interrogate an unknown object. By applying fast neutron imaging techniques, it is possible to estimate the materials (not isotopic composition) and geometry in the object. Using the estimated materials and geometry, components suspected of being enriched uranium are modeled with several enrichments, and simulations predict the time correlations observed by plastic scintillation detectors. The enrichment of the uranium is estimated by comparing the measured time correlation data to the simulated data for each enrichment and choosing the best match.

The method was demonstrated with measurements of a 93% enriched storage casting surrounded by different combinations of DU shields. For each combination, the fast neutron imaging techniques provided reasonable estimates of the known geometry and materials. Using the estimated geometry, the storage casting was modeled with several enrichments. The comparison of the measured time correlations to the predicted ones for each shielding combination clearly shows that the enrichment of the casting is greater than 80%. By comparing the total doubles measured to the total doubles predicted from the simulations, the estimated enrichment of the casting is between 82%
and 95% for the shielding combinations considered. Even though the worst estimate differs from the actual enrichment by 11%, the accuracy of the method is likely acceptable for many nonproliferation applications, including arms control and treaty verification where the goal may be simply to identify the presence of HEU.

This approach has two weaknesses. First, analysis of the tomographic images must permit fairly accurate estimates of the materials and geometry present. Again, the estimates of the geometry from the casting measurements were reasonable as the maximum error of the diameters of the casting and shields was 0.59 cm. If the geometry and materials present are known from other means and the only unknown is the enrichment, then estimates from the tomographic images are not needed. Secondly, the simulations must be able to accurately predict the measured time correlations. The enrichment estimates discussed above are based upon measurements and simulations using small plastic scintillation detectors (2.54 x 2.54 x 15.25 cm). Indeed, predicted time correlations from simulations using the actual dimensions and enrichment of the casting agree extremely well with the data from experiments using the small detectors. Measurements were also made using large plastic scintillator detectors (27 x 27 x 9.5 cm) in an effort to maximize the solid angle of the detectors. Unfortunately, simulations with these detectors consistently overpredict the measured data by varying amounts.

Use of the large detectors for determining enrichment should not be abandoned, but further research into modeling the detector response is needed before undertaking any future large-scale testing activities or building a detector system for production use. Indeed, Henkel and Mullens [46] are already endeavoring in such research. One attempt
at improving the response involves adding a time-component to the pulses in the post-processor. In the large detectors, the light produced can take a longer time to reach the PMT than in the small detectors. Rather than converting all the energy deposited within a certain time window into a pulse as is currently done in the post-processor, each deposition may need to be modeled with a rise and decay time before being combined with other depositions. Other ideas involve adding a position-specific efficiency to the detectors.

Even if future research fails to develop a better way to model the response of the large detectors, a detector system comprised of a large array of the small detectors is possible. Although the setup and electronics of such a system may be cumbersome, a large array of small detectors with a large solid angle would permit accurate predictions of measured time correlations. It may also be possible to accurately simulate the response of other intermediate-sized detectors.

In addition to improving the modeling of the large detectors, other practical factors should be considered before implementing the method for production use. First, for measurements involving the DT generator, a total of 16 simulations (one for each neutron cone) are needed for each enrichment modeled. Most of the simulations in this dissertation required between two and five hours of computational time on a 3.4 GHz Xeon processor. So, the computation time is not trivial. It is possible that tailoring the computational methods for this application could reduce the simulation time. It may also be possible to perform the simulations with many graphics processing units (GPUs) and therefore reduce the computational resources needed. Secondly, extracting the
geometry and material information from the tomographic images and building input
decks for the simulations probably will need to be automated for using the method for
arms control and treaty verification. More than likely, a country presenting an item for
inspection will not want the inner details of the item revealed. So, analysts likely will not
be permitted to view the tomographic images nor see the input decks for the simulations.
With proper development, overcoming these practical factors is possible.
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APPENDICES
Appendix A

Comparison of Primary Casting Measurements to Simulations

The proposed method for determining the enrichment of shielded uranium relies heavily on the ability to accurately predict the time correlations observed by the fission radiation detectors. The geometric model for the simulations of the primary casting measurements is similar to the model shown in Figure 4-14 except that the as-built dimensions of the castings and shields are used. This model compares well to the experimental setup shown in Figure 5-2. 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 features of the building where the measurements were performed.

To determine the threshold to use in the PoliMiPP processor, measurements of a bare $^{252}$Cf source are compared to simulations. In this case, the $^{252}$Cf source is positioned in the middle of the fission radiation detector stand. The thresholds on the CFDs and high voltages are the same in the $^{252}$Cf measurement as the casting measurements. However, the threshold used in the post-processor is adjusted until the simulation closely matches the measurement. The 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 A-1. In the plot, the color purple indicates the sum from all eight fission radiation
detectors, whereas the color black only shows the composite from the detectors on the top row of the stand. The simulations have a neutron threshold of 0.65 MeV. Notice that at 29 ns, the simulation for all detectors is approximately 15% higher than the measurement. Thus, the simulations may give higher time correlations than the measurements for high-energy neutrons. The agreement is better for the case with only the top row detectors. In addition, the agreement at longer times is slightly better than for the case with only the top row detectors. At longer times, the detectors observe more neutrons that have scattered off of the floor or other building structures. This scattering is termed room-return, and the top row detectors are less susceptible to the room-return from the floor.
Using the post-processor threshold of 0.65 MeV, the simulations are compared to the measurements in Figure A-2 through Figure A-5 for all casting and shield combinations. Even without any shielding, the simulations overpredict the fission occurring in the castings. As more shielding is added, the overprediction gets worse. Notice in Figure A-5 that the simulation for DU agrees well with the HEU measured data in the range from 30 to 50 ns.

While Figure A-2 through Figure A-5 show the contribution from all fission radiation detectors and all DT pixels, it is important to examine each detector with each pixel to determine if there is a particular problem with one detector or pixel. To help in this investigation, the percent difference between the simulation and measurement for the integrated counts in the range of 35 to 100 ns is given in Figure A-6. Each subplot in Figure A-6 considers a different shield combination for the HEU casting. The vertical axis on the plots shows the fission radiation detector number, and the horizontal axis shows the DT pixel number. The numbering scheme used for the fission radiation detectors is shown in Figure 4-14. The simulations for fission detectors 1 through 4, which are located on the bottom row of the detector stand, typically overpredict the measurements from 50 to 90% or more. Conversely, the top row detectors typically overpredict by 40% or less. In the bare casting case, the simulations underpredict measurements for DT pixels 1 – 4 and 16. 14 MeV neutrons associated with these pixels are not hitting the casting. As shields are added, more 14 MeV neutrons from these pixels hit the shields, and the underprediction is reduced or eliminated.
Figure A-2. Comparison of simulations and measurements for bare castings (Plot includes the contribution from all DT pixels and all fission radiation detectors.)

Figure A-3. Comparison of simulations and measurements for castings with I shield (Plot includes the contribution from all DT pixels and all fission radiation detectors.)
Figure A-4. Comparison of simulations and measurements for castings with IM shields (Plot includes the contribution from all DT pixels and all fission radiation detectors.)

Figure A-5. Comparison of simulations and measurements for castings with IMO shields (Plot includes the contribution from all DT pixels and all fission radiation detectors.)
Figure A-6. Percent difference between simulation and measurement for integrated counts between 35 and 100 ns for the HEU casting. Each shield combination is shown in subplots. The vertical axis shows each fission radiation detector, and the horizontal axis shows each DT pixel.
Since the bottom row detectors have poor agreement and are influenced more by room-return, work has focused on trying to get good agreement with the top row detectors. Figure A-7 through Figure A-10 compare the single distributions using only the top row detectors, and as expected, the simulations still overpredict the measured response. A comparison of the doubles distributions is shown in Figure A-11 for the castings with all three shielding layers present. To be included in the plot, the two detections must occur in the window from 32 to 100 ns after the alpha detection. For the HEU casting, the simulation overpredicts the total double events by 121%. Similarly, the simulation for the DU casting overpredicts the total double events by 59%. The simulations for the cases with less or no shielding all have similar overpredictions.

Figure A-7. Comparison of simulations and measurements for bare castings using only top row detectors (Plot includes the contribution from all DT pixels.)
Figure A-8. Comparison of simulations and measurements for castings with I shield using only top row detectors
(Plot includes the contribution from all DT pixels.)

Figure A-9. Comparison of simulations and measurements for castings with IM shields using only top row detectors
(Plot includes the contribution from all DT pixels.)
Figure A-10. Comparison of simulations and measurements for castings with IMO shields using only top row detectors (Plot includes the contribution from all DT pixels.)

Figure A-11. Comparison of the doubles distribution for the top row detectors with IMO shields (Plot includes all double events detected between 32 and 100 ns after the alpha detection.)
Appendix B

Efforts to Improve Time Correlation Agreement

Since many discrepancies exist between the simulations and measurements for the primary casting measurements, additional research has been performed to identify the cause and to try to find a way to obtain good agreement.

Better Characterization of Neutrons from DT Generator

In the simulations, the direction of the 14 MeV neutrons associated with each pixel is determined by fitting the response of the imaging detectors to each pixel in a void measurement to a Gaussian distribution. Therefore, the neutrons in the simulation travel in circularly symmetric cones from the DT generator. In reality, each cone of neutrons is elliptical in shape. To characterize the cone of neutrons, an experiment was performed where the DT generator was detached from the arm holding the imaging detectors. The height of the DT generator was constant in the experiment, but the imaging detectors were moved to measure the intensity of the beam at different heights. The imaging detectors were on an arc that was ~ 85 cm from generator at the height of the generator. The intensity of the neutrons associated with pixel 8 is shown in Figure B-1, and the cone of neutrons is clearly elliptical instead of circular.

The data from this experiment was used to create more accurate models for the directions of the neutrons emitted from the DT generator. The simulation with the circular cones and the simulation with the elliptical cones are compared in Figure B-2 for...
Figure B-1. Intensity of neutrons associated with pixel 8

Figure B-2. Simulations using different models for the directions of the 14 MeV neutrons
(The plot considers all DT pixels and the top row fission radiation detectors.)
bare HEU casting. As shown, there is no difference between the simulations. Since using models with the elliptical cones is cumbersome, more computationally intensive, and gives the same result, the circular models were retained in the simulations.

**MCNPX-PoliMi**

In April 2012, Pozzi released a patch to MCNPX-2.7.0 that will create the code MCNPX-PoliMi [45]. MCNPX-PoliMi has the ability to use ENDFB-VII cross-section libraries and has new options for determining the energy of induced fission neutrons based on multiplicity. To test the effects of the updated cross sections and multiplicity options, the case of the bare HEU casting with neutrons coming from pixel 10 was considered. Both the measured and simulated results for pixel 10 and the top row fission radiation detectors are shown in Figure B-3. The legend shows the code and primary cross section library used as well as the multiplicity options, which are defined in the manual, for MCNPX-PoliMi. All of the simulations with MCNPX-PoliMi align with each other. Notice that both MCNP-PoliMi and MCNPX-PoliMi produce the same amount of fission gammas (located at ~10 ns on the graph), but MCNPX-PoliMi produces much more fission neutrons. It also agrees worse with the measurement. The cause for the worse agreement is currently unknown.
Measurements at Oak Ridge National Laboratory

Measurements with large detectors in a similar configuration as small detectors shown in Figure 5-15 were planned for the fall of 2012. However, security events during August of 2012 prevented access to Y-12’s Nuclear Detection and Sensor Test Center. Nevertheless, ORNL has a DU storage casting that is within annular canning. If simulations with the large detectors and the DU casting match measurements as well as the case with the small detectors and DU shields, the discrepancies with the primary measurements are likely related to the interrogation with 14 MeV neutrons from the DT generator. If the simulations do not match the measurements well, then there is an overall inadequacy in simulating the response of the large detectors.
The measurements at ORNL took place in January 2013. The two detectors used had dimensions of 25 x 25 x 8 cm and therefore are slightly smaller than the ones used at Y-12. The detectors are placed in a lead sheath that is ¼ in. thick on the front face and ½ in. thick on the back face. To determine the threshold of the detectors, they were placed on the edge of a cart that is identical to the one at the Y-12 test center. The top of the cart was 93 cm above the floor and the $^{252}$Cf source was 124 cm from the detectors. The measured and simulated time of flight spectra are shown in Figure B-4. The simulations were made using MCNPX-PoliMi, and the post processor uses a neutron threshold of 0.65 MeV. The simulation accurately predicts the gamma peak, response from low energy neutrons, and the room return. However, the simulation over predicts the response from high energy neutrons, and the peak is overpredicted by ~ 27%.

![Figure B-4. Time of flight as measured by large detectors at ORNL](image-url)
In the measurement with the DU casting, the large detectors are arranged as shown in Figure B-5. The large detectors are centered on the DU casting and separated by 25 cm. The singles distributions comparing the measurement and simulation are shown on a log plot in Figure B-6 and on a linear plot in Figure B-7. On a log plot, the simulation compares well to the measurement. However, as more clearly shown on the linear plot, the simulation overpredicts the peak by 18%. The doubles distributions are shown in Figure B-8. The plot shows all double events detected in the range of 5 to 60 ns after the $^{252}$Cf spontaneous fission. Consistent with the other measurements with the large detectors, the simulation overpredicts the double events by 37%.

Figure B-5. Arrangement for interrogation of DU casting with $^{252}$Cf at ORNL
Figure B-6. Singles distribution on log plot for DU casting interrogated by $^{252}$Cf at ORNL

Figure B-7. Singles distribution on linear plot for DU casting interrogated by $^{252}$Cf at ORNL
In addition to interrogating the DU casting with $^{252}\text{Cf}$ neutrons, the casting was interrogated with 14 MeV neutrons from a DT generator. For this measurement, the DT generator did not have a Hamamatsu H9500 pixelated PMT attached. Instead, a 2 in. diameter PMT was attached. A mask with a 1 cm diameter opening was placed between the PMT and generator so that only a narrow beam of neutrons going in the forward direction are counted. The void profile as measured by the fan beam imaging detectors is shown in Figure B-9. Imaging detector 19, which corresponds to detector positions 73 – 76, responded abnormally high, and imaging detector 22, which corresponds to positions 85 – 88, responded abnormally low. Nevertheless, the general shape of the profile is clearly visible. The profile is symmetric around detector position 72, and the blue line shows the profile modeled in the simulations.
The DU casting is centered on the fan beam at detector position 64. Therefore, while all of the beam will hit the casting, most of it will hit one side of the casting. The center of the casting is 21.5 cm away from the DT generator. The large detectors are separated by 25 cm and are parallel to the line centering the casting on the fan beam.

The singles and doubles distributions are shown in Figure B-10 and Figure B-11. In the singles distribution, the simulation overpredicts the peak at 13 ns by 30%. From 20 to 45 ns, the simulation matches the measurement. However, from 45 to 150 ns, the simulation underpredicts the response. The doubles distribution includes pairs detected between 9 and 45 ns after the alpha detection. The simulation overpredicts the total pairs by 19%, but the peak at 0 ns is overpredicted by 30%.
Figure B-10. Singles distributions for DU casting interrogated by DT neutrons at ORNL

Figure B-11. Doubles distributions for DU casting interrogated by DT neutrons at ORNL
Regardless of whether $^{252}$Cf neutrons or 14 MeV neutrons are used for the interrogation, the simulations consistently overpredict the measured time correlations.
Appendix C

DSD File Description

This appendix discusses the detector system definition (dsd) file that is used by the DAUI software. A sample file named Cf_32ImagingDets_8FissDets_16DtPix_2012-02-20.dsd.txt is attached to this dissertation. DAUI is very particular on having parenthesis align, so it is best to always modify a sample file for a new measurement.

Each channel on the NMIS input boards is listed in the dsd file as a “PulseInput,” and Figure C-1 shows an example of a PulseInput. Each PulseInput in the file allows the user to specify which channel the input is associated with and the name of the channel that will appear in the DAUI software. After defining the PulseInput, the signals that are contained on each channel must be defined. In Figure C-1, there are two signals on channel 2 each representing a different detector.

For each signal, there are five fields that must be defined. The first field simply supplies the name of the detector. In the example below, the first signal is named id01 since it comes from the first imaging detector. Any name can be given. On the same line as the name, the user must next specify if the signal is a source. In other words, this field specifies whether the signal is coming from the Cf ion chamber or a pixel on the DT generator. If true, the counts from the signal are used to normalize the counts from the other detectors. The second line specifies the timeskew. Due to electronics and the time signals transit cables, the time of arrival of the signal to the computer will not correspond to the exact time a particle was observed by a detector. A Cf time of flight
measurement is used to set the time skews. Using the speed of light and the distance between the Cf ion chamber and the detector, one can easily determine the time when the gamma peaks should arrive following the Cf fission. For example, suppose there is 120 cm between the Cf ion chamber and the detector. In such a case, the gamma-rays should arrive 4 ns following the Cf fission. If the gamma-rays arrive at 10 ns, the timeskew needs to be increased by 6.

The final two fields that must be defined are the EncodedCategories and PulseWidthRange. In the current NMIS system, the EncodedCategories field will be either PIXEL or DETECTOR. If the signal is coming from one of the pixelated alpha detectors, the

<table>
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</tr>
<tr>
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<td>PulseWidthRange( 74 95 )</td>
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</tbody>
</table>

*Figure C-1. Pulse Input Example*
value is PIXEL. Otherwise, the value is DETECTOR. Next, the PulseWidthRange specifies the range of pulse widths that is expected from the signal. As previously discussed, there are several signals on each channel, and the width of the logical pulse is used to identify each one.

Some older dsd files may contain fields named EncodedValues and DelayTime. However, these fields are not necessary and are not used by the data acquisition software.
VITA

Jason Michael Crye was born in 1976 in East Tennessee. In 1999 and 2001, he received B.S. and M.S. degrees in Nuclear Engineering from the University of Tennessee, respectively. After completing the M.S. degree, Jason served on active duty in the U.S. Navy and was assigned to the U.S. Department of Energy’s Naval Reactors headquarters in Washington, D.C. While at Naval Reactors, Jason managed the design and construction of shielding for all the Navy’s nuclear reactors, refueling equipment, and spent fuel shipping containers.

After completing his commitment to the U.S. Navy in 2006, Jason returned to his beloved hills in East Tennessee and is now an associate research and development staff member in the Nuclear Material Detection and Characterization group at Oak Ridge National Laboratory. At Oak Ridge National Laboratory, he conducts research involving neutron interrogation systems and analyzes data from passive radiation detection systems deployed as part of the Department of Energy’s Second Line of Defense Program.

Jason has a lovely bride and three wonderful children.