Used Fuel Storage Monitoring Using Helium-4 Scintillation Fast Neutron Detectors and Neutron Spectral Analysis

Fuel Cycle Research and Development

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**ABSTRACT**

This project aimed to address the technology gap in the reliable methods to verify contents of sealed dry storage casks for spent nuclear fuel. Helium-4 gas scintillation neutron detectors was proposed to be developed into a neutron spectroscopy and imaging system for nuclear security and safeguards applications. \(^{4}\)He gas is significantly more available than \(^{3}\)He gas and it has unique advantages such as low gamma interaction probability and direct detection of fast neutrons. Due to the sheer amount of fissile materials in a single dry cask, there is a significant neutron fluence emitted whose energies range from thermal to uncollided high-energy neutrons suitable for the detection capability of the \(^{4}\)He detectors. Our project demonstrated the first ever neutron spectroscopic capabilities using \(^{4}\)He detectors, explored novel detector design and performed detector characterization. It further qualified the detectors for use in various applications including high-flux environments with significant gamma-fluence. Computer simulations were used to test various large cask measurement system designs and tests, beyond the compact prototype system and laboratory measurements exploring the feasibility of the neutron spectroscopic approach. Although some cask-characterization methods proved unfeasible, alternative characterization methods using the He-4 detectors were explored and generated promising results.

**PROJECT OVERVIEW**

This project aims to address the technology gap in the reliable methods to verify contents of sealed dry storage casks for spent nuclear fuel. Applying helium-4 gas scintillation neutron detectors to this problem space seemed like a possible successful approach. We created neutron spectroscopic capabilities and proposed prototype detector systems for nuclear security and safeguards applications. \(^{4}\)He gas is significantly more available than \(^{3}\)He gas and it has unique advantages such as low gamma interaction probability and direct detection of fast neutrons. Due to the sheer amount of fissile materials in a single dry cask, there is a significant neutron fluence emitted whose energies range from thermal to uncollided high-energy neutrons that can enable signatures for nuclear materials management.

The project was structured around four main project objectives and a larger number of milestones. While all milestones were addressed to various length of involvement, their common success criteria were dissemination of material through conferences and publications and other documentation that allowed for dissemination of the results. Below is the list of project objectives and Milestones followed by an extended narrative centered on each objective.

![Figure 1: The original \(^{4}\)He detector design (left), the active volume is 304 cm\(^3\). An array of \(^{4}\)He detectors (right).](image)


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**Major project objectives**

**Objective 1:** Create a neutron spectrometer using helium-4 detectors through an experimentally validated neutron energy response function and spectral unfolding techniques.

**Objective 2:** Leverage test measurements and a computation library of spent fuel, enabling the analytical design of a cask-measuring prototype system.

**Objective 3:** Construct a prototype instrument and use it to measure spent fuel casks at a commercial spent fuel storage facility.

**Objective 4:** Develop the capability of cask imaging through a combination of neutron and gamma measurements.

**Milestones**

A set of 12 individual milestones was identified at the start of the project and used as a guiding mechanism for different parts of the work under the project.

### PROJECT MILESTONES

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DETAILED PROJECT OBJECTIVES

Objective 1:

Create a neutron spectrometer using helium-4 detectors through an experimentally validated neutron energy response function and spectral unfolding techniques.

Spent nuclear fuel is currently being stored in dry casks as an interim storage solution. Demand for this storage method has increased globally and the IAEA has expressed a need for robust safeguards and verification technologies for ensuring the continuity of knowledge and integrity of radioactive materials inside dry casks. This research supports the development of a fast neutron spectroscopy system using helium-4 gas scintillation fast neutron detectors to convert the neutron spectra into quantifiable signatures. The goal is to use these signatures to identify diversion/removal of fuel.

Neutron spectrometry is a desirable measurement technique for nuclear material verification and safeguards. For example, neutrons produced from spontaneous fissions and (α, n) reactions represent the main contribution of neutrons originating from spent nuclear fuel. They have unique energy distributions [1], and therefore, detecting the variations of the incoming neutron spectra serves a way to identify the spent fuel materials.

A historic review by Brooks and Klein presented seven methods of neutron spectrometry [2], such as kinematic measurements of recoil particles or charged particles released in neutron-induced reactions, time-of-flight (TOF) measurement of neutron velocity, or the mathematical unfolding of the responses of a neutron detector which are neutron energy-dependent, etc. In this work, the aforementioned methods are all employed on the $^4$He fast neutron scintillation detector to develop its application in neutron spectrometry. Through TOF measurement, response matrix construction, and mathematical unfolding algorithm development, the detector showed its potential ability to differentiate various neutron sources and predict corresponding neutron spectra based on the scintillation light outputs.

The detectors that are proposed to be used in this research are $^4$He scintillation detectors. They are filled with high-pressure $^4$He gas, which is used as a fast neutron detection medium. As in Error! Reference source not found., the neutron capture cross-section of $^3$He is compared with the elastic scattering cross-section of $^4$He (natural helium). For low-energy neutrons, $^3$He has a high capture cross-section. However, when using the $^3$He detectors to detect fast neutrons, it is best implemented in conjunction with moderating materials to make use of the high cross-section at lower energies. In comparison, the elastic scattering cross-section of $^4$He is substantially smaller in the low-energy region. However, for fast neutron detection, the $^4$He cross section exhibits a peak that is located at roughly 1 MeV, nicely matching the peak emission of fission neutrons.
In an elastic scattering interaction, energy is transferred from the incoming neutron to a $^4\text{He}$ nucleus. The neutron is not absorbed in this process. It can keep traveling through the medium ($^4\text{He}$ gas) while the direction and speed are changed. The maximum energy transfer from an incoming neutron to a $^4\text{He}$ nucleus is 64% of the neutron’s energy prior to the interaction due to neutron scatter kinematics.

The kinetic energy transferred to the nucleus is sufficient to strip the electrons from the nucleus that then moves as a recoil alpha particle and excites (equation (1)) or ionizes (equation (2)) other helium atoms along its path within the detector volume.

\begin{align}
\alpha + \text{He} &\rightarrow \text{He}^* + \alpha'. \\
\alpha + \text{He} &\rightarrow \text{He}^+ + e^- + \alpha'. \tag{2}
\end{align}

When ionization takes place, the free electrons often have enough energy to induce a secondary ionization as in equation (3).

\begin{equation}
e^- + \text{He} \rightarrow \text{He}^+ + 2e^- \tag{3}
\end{equation}

These free electrons can recombine with the ionized $^4\text{He}$ atoms as shown in equation (4), which will produce additional excited states [44].

\begin{equation}
e^- + \text{He}^+ \rightarrow \text{He}^* \tag{4}
\end{equation}

These excitations will lead to the production of singlet (more probable) or triplet (much less probable) excimer states [45]. The decay of these excimers to the ground state is an important step to produce scintillation photons, which are either shifted in spectrum through wavelength shifting interactions, or directly detected by photomultiplier tubes (PMTs) at either end, as shown in Error! Reference source not found.. The next chapter covers additional details about this scintillation process.
The $^4\text{He}$ detector technology has several advantages over current neutron and gamma-ray detectors used for nuclear security and safeguards. When comparing with the above-mentioned $^3\text{He}$ detector and its potential alternatives, $^4\text{He}$ is much more available and economical than $^3\text{He}$ gas, and has a better ability to retain energy information of the neutron interactions. Due to its low electron density, $^4\text{He}$ has limited sensitivity to gamma-ray radiation, which in combination with pulse shape discrimination results in excellent gamma-ray rejection. Furthermore, gamma-ray interactions deposit lower energies and have lower light yield in gaseous helium scintillation when compared to neutron interactions, which can be utilized for pulse shape discrimination. Finally, the detector’s performance has been shown not to degrade over time due to high intensity radiation, such as that from spent nuclear fuel. Overall, the superior gamma-ray rejection capability and a more rugged design makes $^4\text{He}$ detectors a potentially very useful tool in spent nuclear fuel monitoring and neutron spectroscopy analysis. The detector can take the measured scintillation light as input and through unfolding reproduce the incident neutron spectrum, which serves a way for “fingerprinting” the spent fuel storage casks. In addition, the $^4\text{He}$ detectors allow for coincidence measurements of neutrons and gamma rays, which is well-known in terms of identifying and characterizing nuclear materials. Other related applications in the areas of radiation detection and instrumentation, including fusion diagnostics, nuclear safeguards, and medical physics, can all benefit from the $^4\text{He}$-based neutron detection techniques.

**NEUTRON SOURCE MEASUREMENTS**

Several sources including $^{252}\text{Cf}$, PuBe and a deuterium-deuterium fusion based neutron generator producing 2.45MeV mono-energetic neutrons were measured by the $^4\text{He}$ detectors. The ability to differentiate neutrons of different energies was also demonstrated through a proof-of-concept active interrogation experiment, which imitates cargo-scanning conditions. Generator-produced 2.45 MeV neutrons were used to induce uranium fission from natural uranium samples. Given the higher energies of induced fission neutrons than the interrogating 2.45 MeV neutrons, $^4\text{He}$ detector was able to detect these high-energy neutrons unambiguously, hence revealing the presence of special nuclear materials. Instrumentation calibration of the $^4\text{He}$ detectors was found to be required and performed which improves the efficiency of high energy neutron detection.

![Figure 3 Event filters were applied to remove events with pile-up, spurious triggers, bad baselines, which would have skewed the largest pulse height cut-off to higher values.](image)
Figure 4 Induced fission neutrons (by neutron generator active interrogation on natural U samples) are distinguishable from generator neutrons in pulse height spectrum comparison.

To demonstrate and prepare how PMT-based $^4$He detectors would be used for practical applications, measurements of neutron signals in a strong gamma background were done. $^4$He detectors have excellent gamma discrimination. The usual requirement for gamma ray sensitivity is set at an exposure rate of 10 mR/hr in evaluating different neutron detectors’ gamma rejection ability. The $^4$He detectors were subject to as much as 500 mR/hr and were still able to distinguish the relatively weak neutron signals of $^{252}$Cf and PuBe sources from the strong gamma background.

**DETECTOR RESPONSE MATRIX CHARACTERIZATION**

At the Edwards Accelerator Lab at Ohio University, various neutron sources were used to cover different ranges of neutron energies, for example, the mono-energetic $14.5$ MeV neutrons from a tritium-target and the continuous-energy neutrons from a beryllium-target. A specific calibration of the photomultipliers was used to accommodate neutrons above 10 MeV. This improves the accuracy of pulse height in the construction of a detector response matrix because the full amplitude of scintillation light can be captured without signal saturation.

Figure 5 Schematic of the time-of-flight experiment set up a Ohio University Edwards Accelerator Facility.
Figure 6 $^4$He detector scintillation output (slow components only) as a result of various incident neutron energies.

$^4$He detector specifications and light responses

The $^4$He fast neutron gas scintillation detector is based on neutron elastic scattering of $^4$He nuclei. The kinetic energy transferred to the nucleus is sufficient to strip away the electrons, and the recoiling nucleus (i.e. α particle) will then either excite or ionize other helium atoms along its path within the detector volume. $^4$He is a relatively efficient scintillator which can produce approximately 15,000 scintillation photons per MeV of energy deposition by neutrons. The detector body is made of stainless steel, and the gas pressure of the $^4$He detectors is about 150 bar [3]. Based on the scaling law of range calculation, for a 2 MeV, 4 MeV, and 6 MeV alpha particle, the range in the detector is about 0.03 cm, 0.07 cm, and 0.13 cm respectively, which is much smaller than the radius of the gas chamber (2.2 cm). Scintillation photons are generated from the de-excitation process of the helium excimers, and are collected by two Hamamatsu R580 photomultiplier tubes (PMT), one on each end of the gas chamber (see Figure 7). The scintillation generated from the excitations is approximately 80 nm, whereas the spectral sensitivity of most PMTs is of the order of 200 to 900 nm [4]. Therefore a wavelength shifting (WLS) material is coated on the inner wall of the gas chamber to change the wavelength of the emitted light. The WLS material absorbs the scintillation light coming directly from the helium gas and re-emits it at a higher wavelength, which better matches the range of sensitivity of the PMTs. The two PMTs on the ends of the gas chamber are set up in coincidence mode so as to suppress the recording of PMT noise signals. PMT calibration was performed to ensure the two PMTs have matching gain factors and the amplified scintillation signals do not exceed the limited dynamic range of the digitizer [5].
Figure 7: $^4$He fast neutron detector. The active volume at the center is pressurized to 150 bar.

The scintillation light can be separated into two components: a fast component which lasts on the scale of tens of nanoseconds and a slow component with a much longer de-excitation time scale of microseconds (see Figure 8). The fast component consists a sharp light pulse which results from the decay process of the singlet excimer state, while the slow component consists multiple short pulses which are produced during the decay process of the triplet excimer state. Due to its low electron density and low ionization density of recoil Compton electrons, $^4$He exhibits excellent gamma rejection and an overall lower light yield (i.e. smaller amount of slow component in scintillation) for a gamma event [6].

Figure 8: Typical digitized PMT outputs of a gamma (left) and neutron (right) event. Fast component lasts approximately 50 ns, followed immediately by a slow component till the end of the 4.27 $\mu$s event window.

Previous results show that the number of photoelectrons detected in the slow component is approximately a factor 3–4 larger for neutron events [6], providing the detector an excellent pulse shape discrimination (PSD) ability. Figure 9 shows a scatter plot of fast component against slow component of the TOF measurement. Events above the black cut-off line will be regarded as neutrons and used for building the detector response matrix. In addition, pulse filtering algorithms [5] are applied to remove pile-up events (i.e. two events within the same event window) and other unreliable events.
Figure 9: Scatter plot of fast component against slow component of the TOF measurement. The nonlinearities in electronics were reduced due to the low gain settings and the pulse post-processing method until outside of the energy range of the TOF measurement (10 MeV).

The $^4$He detectors have the advantage over current $^3$He-based detectors regarding the ability of retaining neutron energy information. Through elastic scattering, an incident neutron transfers up to 64% of its energy to a recoil $\alpha$ particle depending on the elastic scattering angle distribution due to kinematics. However, the relationship between energy deposition and scintillation light production (as shown in Figure 10) is not linear due to scintillator and electronics effects, which are known as the electronics nonlinearity and scintillator nonlinearity [7]. From previous measurements [6], the curved shape of the neutron bend in Figure 10 is likely caused by nonlinearities in the data acquisition system of large signals such as saturation and after-pulsing, which happen more frequently for neutrons of relative high energy. The underlying relationship between the deposited neutron energy and the scintillation light in terms of the slow component is shown as in Figure 10 as obtained from the same measurement as the response matrix measurement below. An empirical power function fitting is chosen here over the linear function fitting, since the former one holds realities in physics (i.e. zero energy deposition leads to zero production of scintillation light), while the linear function fitting results in a non-zero y-intercept.
Figure 10: Scintillation light output (in terms of the slow component) vs. the deposited energy.

**Time-of-flight Measurements**

The so-called detector response matrix (or response function) can be constructed to map the incident neutron energy to the detector scintillation light output. The incident neutron energy can be determined through TOF measurements where the time \( t \) taken by neutrons to travel a known distance \( L \) is tracked and used to calculate the neutron velocity \( v \) and energy \( E \):

\[
v = \frac{L}{t} \quad \text{and} \quad E = \frac{\text{mass of neutron} \cdot v^2}{2}
\]

(Eq. 1)

The TOF measurements were performed using the Tandem Van de Graaf accelerator at the John E. Edwards accelerator laboratory at Ohio University. The Van de Graaf accelerator can accelerate protons, deuterons, or heavy ions on to various targets, such as \(^{10}\)B, \(^{27}\)Al, and \(^{9}\)Be, producing monoenergetic neutrons as well as neutrons with continuous spectra. In this measurement, a 7.5 MeV deuteron beam was pulsed and bunched by a double klystron buncher to produce pulse widths of \(< 2 \text{ ns} \) at the beryllium metal target at \( 60^\circ \) [8]. Neutrons were produced via Be (d, xn) reactions with a continuous neutron spectrum up to 10 MeV. The generation of neutrons followed the pulse frequency of the deuteron beam, at 1600 ns between pulses. For the chosen flight path this corresponds to the slowest neutrons from each pulse to reach the detector before the next pulse, to be around 200 keV, below which the \(^4\)He (n, elastic) interaction probability is negligible [6]. Associated with the deuteron beam operation, gamma rays are also produced from neutron activation of materials, where the excited nucleus often decays with short half-lives by emitting gamma rays. More details regarding the detector settings and accelerator facility discursions can be found in previous measurement [9].

Gamma rays travel at the speed of light and therefore, as shown in Figure 11, the TOF spectrum has a main peak (so-called gamma flash), followed by neutron events with various energies along the 1600 ns window. The timing resolution can be determined by the FWHM of the gamma flash. It is calculated as 14 ns and can then be used to estimate the energy resolution of the TOF measurement. Due to the relative low sampling resolution (8 ns) of the DRS4 waveform digitizing chip of the data acquisition system [6], interpolation between two adjacent timestamps is chosen over Gaussian fitting which may contain large uncertainties. The relative energy resolution is estimated as 9.5\% at 6 MeV, 5.6\% at 2 MeV, and 4.0\% at 1 MeV, which accounts for the
uncertainties in both neutron flight time and its travel distance. The gamma flash is caused by data acquisition module uncertainties and electronic noises, and is used as the reference timestamp of neutrons originating from the target. From the same deuteron beam pulse, neutrons of different energies are generated and collimated down the TOF tunnel, reaching the detector at different timestamps. The TOF is calculated by subtracting the timestamps of neutron events at the detectors and the gamma flash timestamps.

Figure 11: Measured TOF spectrum. The data shown represents around $6 \times 10^6$ events.

The target used in this work was $^9$Be, which produces neutrons of energies between 50 keV and 10 MeV. Figure 12 shows the expected incident neutron flux (integrated over the detector solid angle) from the $^9$Be (d, n) reaction. The expected neutron flux is determined in previous work [10], using existing calibrated neutron detectors at the accelerator facility.

Figure 12: Experimental neutron flux from the $^9$Be (d, n) reaction.
By calculating the ratio between the number of detected neutrons and expected neutrons as in Figure 12, the energy-dependent efficiency is shown as in Figure 13. The maximum efficiency was 6.8% at 1.5 MeV, due to the large neutron scatter cross section for $^4\text{He}$ around that energy [6]. The bump in the efficiency curve around 3 MeV may arise from the fluctuations which were measured by the calibrated detectors as well (see Figure 12). In addition, the efficiency obtained from MCNPX PoliMi Monte-Carlo simulation is plotted and compared with the measured efficiency. A beam of 20 groups of monoenergetic neutrons ranging from 0.5 MeV to 10 MeV (increased by 0.5 MeV) are simulated, where neutrons will travel 10 m in air before hitting the detector. At 150 bar, the density of $^4\text{He}$ gas is about 0.02464 g/cm$^3$, and default MODE: N is used during simulation. The nuclear data library ENDF/B-VI is chosen for neutron evaluations up to 20 MeV, which contains information on neutron collisions, such as cross-sections and outgoing neutron energy [11]. Elastic scattering reactions only result in energy transfer without any other secondary effects [12]. Moreover, in this simple check, we are only concerned about the numbers of neutrons interacting with $^4\text{He}$ nuclei and their energy deposition (used for the construction of the kinetic response matrix), therefore it is acceptable that the emission data for charged particles or recoil nuclei transport is not simulated or tracked here. The output from simulation contains statistical errors corresponding to one standard deviation [13], and the uncertainties in measured efficiency are calculated via error propagation, which result from the uncertainties in the expected neutron flux. Both curves have a peak around 1-2 MeV, while the simulated one has higher efficiency values below 2 MeV. It may result from the conservative PSD algorithm, which would sacrifice a considerable amount of low energy neutrons due to their relative small pulses. In addition, the simulation simplified the geometry configurations by assuming air only between the neutron source and the detector, while in reality, neutrons may undergo scattering within the collimator or tunnel. The relative large differences between measured and simulated efficiency agree with previous study [9], at 1 MeV, approximately the measured detection efficiency was 6.2% while the simulation predicted 11%.

Figure 13: Black line: $^4\text{He}$ detector intrinsic efficiency as a function of incident neutron energy from TOF measurement. Red dash line: $^4\text{He}$ detector intrinsic efficiency as a function of incident neutron energy from MCNPX.
Detector response matrix

The detector response matrix is constructed on a 2-D histogram where the two base axes are the incident neutron energy and the detector scintillation light outputs. Figure 14 illustrates the relationship in terms of either total scintillation light output (i.e., slow component plus fast component) or just slow component only. Large TOF values correspond to low energy neutrons, and as expected, low energy neutrons tend to produce low scintillation light output.

![Figure 14: Slow component only vs. TOF (left), and total scintillation light vs. TOF (right). Visually, the addition of fast component to slow component (i.e. total scintillation light outputs) does not noticeably improve the relationship.](image)

It was reported in [6] and confirmed in Figure 14 that the distribution of slow component values is related to the neutron energies. Additionally, Ting [5] addressed that the fast components of the detector scintillation light may introduce large uncertainties in the pulse integration. Fast components have short time scales on the order of nanoseconds and tend to exceed the dynamic range of the digitizer, depending on both gain settings and the radiation intensity. Therefore, the response matrix is constructed by using slow component values only. Each neutron event can be defined by its initial energy which is calculated from its TOF, and by the scintillation light (slow component) it produced, which is measured by the $^4$He detector. The slow component is divided into 1000 bins to reduce the statistical fluctuations since even small variations in the measured light output distribution, such as interfering background radiation, will result in wide fluctuations which can be observed in the unfolded neutron spectrum. The incident energy is divided into 20 bins (0.5 MeV bin width) to account for the detector's energy resolution (in another word, timing). All the events are then grouped into corresponding bins which creates a bivariate histogram of 1000-by-20 bins with normalized counts. It describes the unnormalized distribution function of events as a function of the energy ($E$) and slow component of the scintillation light ($L$) as $R(L, E)$. The final detector response matrix is given in Figure 15.
In addition to the TOF measurement, the detector response matrix can also be obtained from MCNPX-PoliMi simulations. The same simulation model is used as in the efficiency simulation. The deposited energy in one elastic scattering reaction is a function of scatter angle and is calculated as the difference between the incoming and outgoing neutron energy. For each monoenergetic neutron group, the distribution of energy deposition is plotted with 0.5 MeV bin width. By combining all groups of neutrons, the kinetic response matrix is built, which shows a probabilistic mapping between the incident neutron energy and the deposited neutron energy as in Figure 16 (left) as the first step. The deposited energy can then be converted to scintillation light by the relationship as shown in Figure 10, and the final simulated response matrix is shown in Figure 16 (right). The simulated response matrix has the same diagonal features as the measured response matrix and tends to yield larger scintillation light output along the whole energy range. The reason is that in real measurements, due to the effect of photon-statistics and detectors resolution, the scintillation light output distributions will be broadened and smoothed. In the simulations, the resolution of the detector has to be based on empirical data [6], and has not yet been added to the simulation process because we are not able to quantify it at this point, thus the simulated response matrix represents an ideal light distribution. Nonetheless, it is a rough comparison and serves as a verification of the measured response matrix, therefore the following unfolding process will still use the measured response matrix.

Figure 15: Response matrix for $^4$He detector from TOF measurement.

Figure 16: The simulated neutron energy-deposition distribution (left), and the final response matrix for the $^4$He detector from MCNPX simulation (right).
SPECTRUM UNFOLDING METHOD

Spectrum unfolding is a de-convolution optimization process, utilizing both the detection response function and the measured scintillation light distribution.

The relationship between the incident neutron spectrum \( \phi(E_n) \) and the resulting light output spectrum \( N(L) \) in a scintillator can be written as follows [14]:

\[
N(L) = \int R(L, E_n) \phi(E_n) dE_n \quad \text{(Eq. 2)}
\]

\( L \) is the corresponding scintillation light output produced by an incident neutron with energy \( E_n \). \( R(L, E_n) \) is the detector response function, which represents a relationship between the neutron spectrum and the light output spectrum. When given a well-constructed \( R(L, E_n) \), a good estimation of \( \phi(E_n) \) can be obtained. However, the resolution of \( \phi(E_n) \) is limited by discrete binning of \( R(L, E_n) \). Eq.3 shows the matrix form of Eq.2, with \( K \) groups discrete intervals of energy \( E \), and \( M \) groups of light output \( N(L) \).

\[
\begin{bmatrix}
N_1 \\
\vdots \\
N_M
\end{bmatrix} =
\begin{bmatrix}
R_{1,1} & \cdots & R_{1,K} \\
\vdots & \ddots & \vdots \\
R_{M,1} & \cdots & R_{M,K}
\end{bmatrix}
\times
\begin{bmatrix}
\phi_1 \\
\vdots \\
\phi_K
\end{bmatrix}
\quad \text{(Eq. 3)}
\]

The bin width of the light output can be adjusted. Lower light outputs have narrower bin width than relatively high light outputs in order to account for statistics. Energy bins are set as 0.5 MeV wide. Each event from the TOF measurements is sorted into the corresponding energy and light output bin.

Iterative Least Squares Fitting

The basic solution to the matrix inverse problem can be obtained by commonly known computational algorithms. However, the direct inversion can produce negative solutions of flux, which are nonphysical in our unfolding problem of real neutron spectrum. Therefore, it becomes necessary to develop a constrained optimization method.

Matlab can solve least squares problems with bounds or linear constraints, providing a solution that will be regarded as an initial “guess spectrum”. The “guess spectrum” is non-negative and obtained directly from matrix inversion. Upon which, the iterative Least Square Method (LSM) algorithm [15] will be applied to find the best \( \phi_j \) by iteratively performing the following:

1. A reasonable “guess spectrum” \( \phi_j \) is found.
2. Set \( j = 1 \). Calculate the value of Eq.4 below, using \( \phi_j \) from step 1. \( W_i \) are chosen to be the square roots of the counts of each light output bin, which are inversely proportional to the uncertainties of the counts.

\[
f(\phi) = \sum_{i=1}^{M} w_i (N_i - \sum_{j=1}^{K} R_{i,j} \phi_j)^2
\quad \text{(Eq. 4)}
\]

3. Determine whether or not the value of \( f(\phi) \) can be reduced by increasing or decreasing \( \phi_j \).
4. If the result of (2) cannot be further reduced, or \( \phi_j \) is less than 0, set \( j = j + 1 \).
5. Repeat (2), (3), and (4) until \( j = K \).

The solution of \( \phi_j \) will eventually converge to a most optimal solution that is both positive and having the minimized \( f(\phi) \). This method uses the solution of the matrix direct inversion as a first
“guess”, then optimizes it via the iterative improvement-based quadrature method which is widely known for solving ill-conditioned problems [16].

**Uncertainty Estimation**

The uncertainties in the unfolded spectra could come from both the measured response matrix \( R(L, E_n) \) and the light output spectrum \( N(L) \). The uncertainties associated with \( N(L) \) are from detector readings and can be determined by Poisson statistics [17]. While the uncertainties in the response matrix have to be either calculated from error propagations, which requires matrix derivatives [18] and the knowledge of the uncertainty matrix (covariance matrix) [17], or computed from existing unfolding codes such as MAXED [19] and FERDO [20].

In this work, we applied a stochastic method in order to overcome the lack of the detectors energy resolution and the unavailability of a priori information of the response matrix and particle fluence in the above methods. When building the response matrix, neutrons are sorted into corresponding energy and light output bins, and their energies are calculated based on the TOF. Therefore, uncertainties in the response matrix are in part caused by timing uncertainties of the TOF measurement. Previous work [21] shows that the \(^4\)He detector has a 3.229 ns full width at half maximum (FWHM) time resolution (modeled by Gaussian) at similar detector settings as the TOF measurement. Consequently, a time spread following a Gaussian distribution with 3.229 ns FWHM is randomly added to each TOF value. The uncertainties of the counts in each light output bins are added to \( N(L) \) in the same way based on the assumption of Poisson statistics in the nuclear detection counting system [22]. 10,000 trials were conducted, resulting 10,000 response matrices therefore 10,000 unfolding results. The maximum and minimum values reflect around +/- 3 sigma, where about 99.7% values will lie within the estimated bound [23].

**UNFOLDING RESULTS**

In this paper, three measurements were conducted with a \(^{252}\)Cf spontaneous fission neutron source, a PuBe (\(\alpha, n\)) neutron source, and a deuterium-deuterium (D-D) fusion-based neutron generator to test the iterative LSM with the experimentally measured response matrix.

The sample data is collected separately by using a 115 \(\mu\)Ci \(^{252}\)Cf source and a 10 Ci PuBe source, which are placed at 35 cm from the center of the detector's active volume. Within 2 minutes, a total of \(5.02 \times 10^4\) and \(1.11 \times 10^5\) counts are collected from the \(^{252}\)Cf source and PuBe source respectively. Neutrons are monoenergetic of 2.45 MeV from the University of Florida's D-D neutron generator. The detector is placed 1 m from the generator, and \(7.65 \times 10^4\) counts are collected during 2 minutes. Same gain setting (a relatively small gain was used to avoid saturation) and pulses filters are applied as the TOF measurement at Ohio University to keep the consistency. All the measurements are taken at room temperature. No additional gamma-ray shielding materials are used during the measurements due to the gamma-ray insensitivity of the detectors.

Figure 17 shows the light output spectra after pulse shape discrimination. The D-D monoenergetic 2.5 MeV neutron source produces overall the smallest amount of scintillation light, while for the (\(\alpha, n\)) and spontaneous neutron source, it is not possible to gain much information from the detector pulse height spectrum directly, yet the unfolded neutron spectra revealed unique features as shown in Figure 18. As mentioned above, no a-priori information about the incident neutron spectrum is required, which mimics the blind-measuring case during nuclear materials monitoring. In the spectrum unfolding, non-linear bin width was chosen to lower the impact of the statistical fluctuations especially for large scintillation light amplitudes. More fluctuations are observed after 30,000 (arbitrary units (a.u.)) in the light output spectra, but they only account for
6% of the total events. Overall, the unfolded $^{252}$Cf spectrum exhibits a factor of 1.38 discrepancy from the Watt distribution of a typical spontaneous fission source [24], which has a maximum probability around 1 MeV and decreases gradually afterwards. These characteristics can be used for distinguishing from ($\alpha$, n) neutrons, which usually have a higher yield between 2.5 MeV and 3 MeV based on previous work using simulations [25] as well as shown in the unfolded spectrum. Information for the specific PuBe source is limited, therefore we are not able to plot a reference spectrum. The unfolded spectrum of the D-D measurement has a peak at 2.5 MeV with a relatively small uncertainty. 53% of the neutrons are lower than 2.5 MeV, which may have resulted from some of the neutrons being slowed down by scatter and room return. And there is only a minor predicted fraction of neutrons with energy higher than 3 MeV which agrees with the expectation. In addition, there is another way to evaluate the unfolded spectra in terms of their average energy and high energy neutron ratio. The PuBe unfolded spectrum yields an average energy of 4.35 MeV, and 50.77% neutrons are higher than 4 MeV. While the $^{252}$Cf unfolded spectrum has a 2.63 MeV average energy, and 21.91% neutrons are higher than 4 MeV. These features provide a proof-of-concept to identify and verify various neutron source types. For example for nuclear spent fuel monitoring, where the spontaneous fission neutrons and the ($\alpha$, n) neutrons have their unique contributions as a function of burnup and cooling time to the overall neutron emission spectrum [25].

Figure 17: The light output spectra of the three measurements.
Figure 18: The unfolding results (left), and a zoom in of the unfolded spectra (right), of the three measurements. The error bars reflect approximately 3 standard deviations.

\textbf{\textsuperscript{4}He DETECTOR SPECTROSCOPY CONCLUSIONS}

These results present the characterization of \textsuperscript{4}He neutron detectors and their potential application as a neutron spectrometer. An experimentally determined neutron response function from the TOF experiments was used and an iterative least squares unfolding algorithm was developed to obtain the neutron spectra from a \textsuperscript{252}Cf spontaneous fission source, a high-energy PuBe (\(\alpha, n\)) neutron source, and a 2.45 MeV monoenergetic D-D neutron generator. Notable discrepancies and uncertainties in the unfolded spectra were found, yet expected characteristics for different types of neutron sources were observed and easily distinguished. The recoil spectrum unfolding is unlikely to match the incoming spectrum as perfectly as certain detectors designed for spectroscopy applications [26], but it has the benefit of directly using the existing detectors without complex electronics. In this work we are focusing on differentiating various neutron sources for spent fuel monitoring purpose, where the main contributions of neutrons are from spontaneous fissions and (\(\alpha, n\)) reactions. While separate neutron sources looked significantly different, the results and discrepancy of unfolded spectra show that mixed neutron sources of small difference would be very hard to differentiate using the detectors used here. The results addressed the advantages of the novel \textsuperscript{4}He fast neutron scintillation detectors and supported the proof-of-concept idea of using the detectors to verify the content of the spent fuel dry casks. Other applications in the areas of nuclear nonproliferation and homeland security can also benefit from this work.

Future work could include the comparison of the iterative LSM with other neutron spectrum unfolding codes such as FERDO [20] and GRAVEL [27], multi-source measurement and unfolding, and ultimately, the development of neutron analysis system with quantifiable signatures outputs for spent nuclear fuel monitoring [28].

\textbf{PSD Methods for PMT-based \textsuperscript{4}He Detectors}

Numerous techniques have been developed to separate the digital signals produced by neutrons from those produced by photons based on the fundamental difference between the two signals, which is mainly the time characteristics of the pulses while they decay. These methods include the charge comparison method (CC), pulse gradient method (PGA), and simple digital charge comparison method (SDCC), which are the three methods investigated below.
Charge Comparison (CC) Method

The CC method compares the integral of the fast and slow components of each pulse (see 19) by plotting them on the x and y axis, respectively [29]. The integration window for each component was varied until the separation was not improving anymore. For He-4 pulses with 8.33 ns window between consecutive points, the optimum integration window for the fast component was found to be 25 ns before and 17 ns after the pulse peak location. The corresponding slow component integration window starts at 8 ns after the peak location and goes all the way to the end point.

The code was constructed in a way that allows semi-automatic generation of the separation line to be used for classifying signals in to neutrons and gammas (see Error! Reference source not found.). The object “PSD_class”, in which all the three separation algorithms are defined, is initiated with three sets of data. The “Test_data_1” and “Test_data_2” represent the signals from pure gamma source (60Co or 137Cs) and a mixed source of neutron and gamma (252Cf), respectively. When calling any of the three separation methods, a PSD plot is generated using these two test datasets, where the user is able to make as many clicks as desired on the plot to guide the separation line. These points are shown as red dots in Fig. 19 a. The plot window records the location of these points and creates the separation line that is a polynomial fit of a given order. Two more plots (scatter and density plots) are then generated showing the performance of the line at separating the signals in the “Final_data” in to neutrons and gammas. The entire process is shown for constructing the CC method in Error! Reference source not found..

In [1]: ‘’’ Running the PSD code ‘’’
   ...: PSD_Test = PSD_class(Final_data, Test_data_1, Test_data_2)

Figure 19.– Running the PSD code

Figure 20. He-4 signals: Constructing the CC method using 252Cf (blue) and 60Co (green) signals (a), testing the decision line on separating 252Cf signals in to neutrons (red) and gammas (green) (b), and the corresponding density plot (c).

Pulse Gradient Analysis (PGA) method

The PGA method differentiates between neutrons and gammas using the ratio between a data point in the decaying region of the pulse and the maximum amplitude [30]. Signals produced by photon impact decay faster than those produced by neutrons, so the ratio is expected to be larger for neutron signals given that an optimum time window is chosen between the two points.
Figure 21. Sample $^{252}$Cf and $^{60}$Co signals recorded with He-4 detector (a), modeling the slow components with 4th order polynomial for improved PSD performance by the PGA method (b).

However, as shown in Error! Reference source not found., since the pulse decay does not have a smooth profile for signals produced by He-4 detector, it was impossible to achieve any separation between neutrons and gammas using this method. The solution was modeling the slow component by a smooth function (Error! Reference source not found. b) and then choosing an optimum location for the second amplitude, where the two models are separated the most. Selecting only the slow component of the pulse for the polynomial fit and following a vectorized approach using “numpy” package in Python made the additional time required by the fitting step insignificant. The optimum location for the second amplitude was found to be 0.5 μs after the main peak location.

The process of constructing the separation line and testing it with $^{252}$Cf signals is shown in Fig. 21, which is the same process explained for the CC method previously.

Figure 22. He-4 signals: Constructing the PGA method using $^{252}$Cf (blue) and $^{60}$Co (green) signals (a), testing the decision line on separating $^{252}$Cf signals into neutrons (red) and gammas (green), and the corresponding density plot (c).

Simple Digital Charge Comparison (SDCC) method

Separation using the SDCC method is based on integrating the square of the slow component and passing it through a log function, which yields a separation parameter called “D parameter” [30]. The formula for calculating the parameter is shown in Eq. (1).
\[ D = \log \left( \sum_{n=a}^{n=b} x_n^2 \right) \] (1)

Figure 23. He-4 signals: Constructing the SDCC method using \( {}^{252}\text{Cf} \) (blue) and \( {}^{60}\text{Co} \) (green) signals (a), testing the decision line on separating \( {}^{252}\text{Cf} \) signals into neutrons (red) and gammas (green) (b), and the corresponding density plot (c).

The optimum integration window for calculating the separation parameter in the case of SDCC was 25 ns after the main peak location and goes all the way to the end point. As a result, \( a \) and \( b \) in Eq. (1) represent 8 ns after the pulse amplitude and the last pulse data point, respectively. The process of constructing the separation line for this method and testing it with \( {}^{252}\text{Cf} \) signals is shown in Fig 22.

In the next step, using the three PSD algorithms constructed, optimization of separation between neutron and gamma signals was repeated for Ej-309. For the CC method, the optimum integration window for the fast component was found to be 12 ns before the pulse peak location and 32 ns after. The corresponding slow component integration window starts 32 ns after the peak and extends until the last data point. For the PGA method, the optimum location for the second peak is 60 ns after the main peak location. The optimum integration window for calculating the separation parameter for the case of SDCC was 40 ns after the main peak location and goes all the way to the end point. Besides varying the integration window for the different separation parameters, the only difference in the case of Ej-309 was that \( {}^{137}\text{Cs} \) was used as the pure gamma source, instead of \( {}^{60}\text{Co} \), since it was already used during the energy calibration step. The performance of separating \( {}^{252}\text{Cf} \) signals into neutrons and gammas using Ej-309 detector and the three separation algorithms is shown in Fig. 23.
Figure 24. Ej-309 signals: Separating $^{252}$Cf signals into neutrons (red) and gammas (green) using the CC method (a), PGA method (b) and SDCC method (c)

**PULSE SHAPE DISCRIMINATION OPTIONS FOR SIPM-BASED $^4$He DETECTORS**

The focus of the PSD was to evaluate the effectiveness of TOT (Time-over-threshold), Arktis’ specified PSD method, and look at other pulse discriminating qualities to distinguish gammas and neutrons. Fig. 24 shows a 100-pulse example of Cf-252 measured at the Sandia National Laboratory; various threshold lines have been added to the figure.

If the threshold is too low, the discrimination ability of TOT for gamma and neutron events is limited. Using an extremely high-value above baseline thresholds, such as at 10000 digitizer units, does two critical things: it removes gamma events almost completely from the data and the TOT distribution will artificially decrease which makes any remaining gamma events difficult to process.

Initially, TOT for all individual pulses without using a 30 ns coincident gate window was evaluated. Figure 26 shows Cf-252, AmLi, AmBe, Co-60, and a background measurement from Sandia National Laboratory using threshold 6500. The majority of Co-60 has a lower TOT distribution than the neutron sources. This indicates that lower TOTs, if the threshold setting is optimized, are more likely to be gamma events than higher TOTs.
However, there is a secondary Co-60 TOT peak that corresponds to the background TOT distribution. This indicates the helium-4 SiPM detector is sensitive to higher energy events such as cosmic radiation. The SiPM’s ability to detect background neutrons and higher energy events was further examined at the University of Florida.

![Figure 26. Data collected at Sandia National Laboratory showing the effects on TOT for different sources.](image)

Although there is promising distinction between TOT for individual pulses collected from gamma and neutron sources, Arktis specifies that they use a 30 ns gate window for their TOT evaluation. As long as two or more events are above the threshold value within a 30 ns window, then TOT is measured. When this 30 ns gate window was applied, pulses that were within the 30 ns gate window were grouped together and are referred to as groups of “multiples.” Pulses that were not within 30 ns of another pulse are referred to as groups of “singles.”

Overall, the neutron sources had higher percentages of groups of “multiples,” ranging between 37-49% compared to the Co-60 source. In contrast, Co-60, a gamma source, only 12% are groups of “multiples.” 88% of Co-60 was found to be in groups of “singles.” This shows that the 30 ns gate window discriminates between neutron and gamma sources. Furthermore, 51% of the background pulses are in groups of “multiples” which indicate they are higher energy events. This supports the theory that the majority of the background data were from cosmic rays.

Analysis of the scintillation pulses in each segment for TOT, peak amplitude, and integral showed minor discrepancies among the three detector segments. For example, a Cf-252 source was positioned 0.4 m from the center of detector’s segment 2. The distributions of the TOT values for SiPM pulses for each segment are shown in Figure 27. It was expected and confirmed that segment 2 had a greater amount of measured events.
This discrepancy between segments 1 and 3 is also observed in Figure 28, which show the distributions of the values of SiPM pulse peak amplitude and pulse integrated area in each segment.

Overall, Figs. 26-27, indicate that either there may be some inconsistency of scintillation characteristics among the three segments of the detector, which are supposedly optically independent and identical. The last step in characterization is the choice of optimum performance region for each PSD method to be followed by combining them all according to Figure 25.
**Objective 2:**

*Leverage test measurements and a computation library of spent fuel, enabling the analytical design of a cask-measuring prototype system.*

**COMPUTATIONAL METHODOLOGY DEVELOPMENT**

Computational methods include use of the NGSI Spent Fuel Library, ORIGEN-S, MCNP, and creation of MATLAB linkage code. The NGSI Spent Fuel Library addresses the geometry and composition of spent fuel assemblies. ORIGEN-S is used to generate neutron source terms using decay data. MCNP is used to simulate the neutron transport through the fuel assemblies and cask system. The MATLAB linkage code is used to transfer material composition data from the NGSI spent fuel library to ORIGEN-S for neutron source term generation, and then use MCNP for transport and shielding calculations.

**ORIGEN-S**

The neutron source spectra and intensities are calculated using ORIGEN-S based on the known compositions of assemblies from the NGSI spent fuel libraries. ORIGEN-S quickly produces a neutron source term for a given mass and composition of spent fuel using standardized decay data. The neutron source calculated by ORIGEN-S includes neutrons produced from both spontaneous fission and \((\alpha, n)\) reactions originating from the decay of to heavy nuclides. The alpha particle is emitted from the decay of a heavier element and can initiate a nuclear reaction in lighter elements, such as the transformation of oxygen to neon, while emitting a neutron. The neutrons from photofission and photoneutron reactions are excluded since their intensity is minimal in the case of spent fuel. The typical minimal threshold energy for a photonuclear reaction, about 8 MeV, is much higher than the typical energy of gamma rays emitted from radioactive nuclides in spent fuel [53].

ORIGEN-S input files have an 80-character line limit and very little non-numerical text. The program is controlled by “vectors,” or one-dimensional arrays that specify the amount of each nuclide being considered. ORIGEN-ARP is a graphical user interface (GUI) that translates the entered data into a valid ORIGEN-S input file. To calculate neutron source spectrum information, ORIGEN-S requires isotope masses in grams. The timeframe was configurable and to enable normalization of results it was specified to one second. Due to the large amount of data involved, this was not feasible to input using ORIGEN-ARP. A later section discusses the development of a MATLAB linkage code that automated creation of the ORIGEN-S input files.

**MCNP**

MCNP is used to transport the neutron source spectra through the fuel assemblies and cask system. The large size of the cask (approximately 3.3 meters in diameter and 6 meters tall) presents a considerable challenge to model in MCNP. Transporting neutrons through strong shields such as those found in a dry storage cask results in significant changes in neutron energy and direction due to absorption and scattering. MCNP generates a new random number each time a particle crosses a cell boundary, which increase computational timeframes for complicated geometries such as a cask.

MCNP defines cell volumes in terms of surfaces and Boolean logic. MCNP uses material cards to generate a homogeneous fill material at a specific density to occupy cells. The weight percent of each isotope is defined within each material card. This is done using the isotope ZAID number, which contains six digits \(ZZZAAA\), where \(ZZZ\) is the atomic number \(Z\) and \(AAA\) is the atomic
mass number A. For example, $^{235}$U has the ZAID number 092235. This allows MCNP to reference given cross section data for the isotope in the ENDF/B-VII database.

Various types of tallies are used to track information of interest about the particles as they pass through the defined geometry. All tallies are normalized to one source particle. This research utilized the average surface flux tally (F2) and the average cell flux tally (F4) to track the number and energy of neutrons. Each tally is separated into different energy bins and recorded with units of particles per square centimeter. Due to the earlier normalization in ORIGEN-S source term generation to one second, this was easily converted into a neutron flux.

**MATLAB Linkage Code**

One of the goals of this work is to develop accurate transported neutron spectra for spent fuel inside a dry storage cask. This required creation of a MATLAB linkage code since ORIGEN-S and MCNP use completely different formats for inputting and outputting data. Figure 29 shows the steps necessary to move between programs and obtain useful results.

Step 1: NGSI Spent Fuel Library. The NGSI spent fuel libraries are in MCNP format with detailed geometry for the entire assembly and material compositions for each fuel rod. To generate a neutron spectrum, this information must be processed into an ORIGEN-S input file. This was completed using a similar methodology as the General Automated Transition of Required data for SOURCES (GATORS).

Step 2: Extract Data. Running an MCNP input file in initialization mode with the table option in the command line prints relevant tables needed. Print table 40 contains the mass fraction information for each isotope. Table 1 Partial Table 40 from MCNP initialization from 4% initial enrichment, 15 GWd/MTh burnup and 5-year cooling time NGSI spent fuel assembly shows a partial table 40. Print table 50 contains the total mass for each cell. Table 2 shows a partial table 50. Multiplying the mass fraction and the total mass for each cell computes cell-based isotope masses in grams, which is required for the ORIGEN-S input file. A bash script was used to execute MCNP using only the initialization option for all files in the NGSI spent fuel library to generate the necessary data tables.

Step 3: ORIGEN-S Input File. The MATLAB linkage code was used to read the MCNP input files that now contained table 40 and 50. This script then built ORIGEN-S input files using a template and inserting relevant data from each rod. Documentation on the ORIGEN-S input format is available from Crabbs [31].

Step 4: Execute ORIGEN-S. ORIGEN-S was executed for each rod using a bash script.

Step 5: Neutron Spectra. The ORIGEN-S output files present details for all neutron source activity in each rod.

Step 6: MCNP Input File. A MATLAB script converted the neutron source spectra into MCNP format and built an updated MCNP input file.

Step 7: Execute MCNP. MCNP was executed to transport the neutron spectra and collect data using tallies.

Step 8 Transported Spectra. The final MCNP output file contains neutron energy spectral data that is analyzed to provide useful results.
**Errors and Uncertainties**

Computer simulation models imply uncertainties from inherent approximations and from input parameters, such as cross sections [32]. Uncertainty in calculations is due to the uncertainty in physics data and simplifications in modeling techniques [33]. Uncertainties begin in the initial isotope compositions from the NGSI spent fuel libraries. Upon carrying these isotopic compositions to ORIGEN-S the next variable is the uncertainties in decay constants and fission spectra. Then MCNP has uncertainties in the cross sections as the neutrons are transported through the geometry. Uncertainties in decay constants, cross sections, and fission spectra lead to further uncertainties in the final neutron flux and neutron energy spectrum measured in the tally results. Evaluating the propagation and impact of these uncertainties is the subject of ongoing research [34][35].

Another way to evaluate errors and uncertainties is to make comparisons between calculations and experimental isotopic measurement data. Radiochemical isotope assay data for PWR assemblies from the Takahama-3 reactor indicate agreement within a few percent of code predictions of SCALE 4.4 [36].

The precision of MCNP results depends on the number of histories run. MCNP tallies are normalized to the number of starting particles and printed with the relative error. For F2 and F4 tallies, acceptable values of relative error should be less than 10%. MCNP also performs 10 statistical checks for the estimated answer for the tally fluctuation chart bin. These statistical checks evaluate aspects of the tally results including mean behavior, changes in relative error, variance of the variance, figure of merit, and the probability distribution function’s slope.

*Table 1* Partial Table 40 from MCNP initialization from 4% initial enrichment, 15 GWd/MTU burnup and 5-year cooling time NGSI spent fuel assembly

<table>
<thead>
<tr>
<th>Material Number</th>
<th>Isotope</th>
<th>Mass fraction</th>
</tr>
</thead>
<tbody>
<tr>
<td>51</td>
<td>92234</td>
<td>2.14505E-04</td>
</tr>
<tr>
<td></td>
<td>92235</td>
<td>2.26959E-02</td>
</tr>
<tr>
<td></td>
<td>92236</td>
<td>2.34057E-03</td>
</tr>
<tr>
<td></td>
<td>92238</td>
<td>8.38138E-01</td>
</tr>
<tr>
<td></td>
<td>93237</td>
<td>1.28933E-04</td>
</tr>
<tr>
<td></td>
<td>94238</td>
<td>1.41305E-05</td>
</tr>
<tr>
<td></td>
<td>94239</td>
<td>4.09108E-03</td>
</tr>
<tr>
<td></td>
<td>94240</td>
<td>7.30120E-04</td>
</tr>
<tr>
<td></td>
<td>94241</td>
<td>2.92078E-04</td>
</tr>
<tr>
<td></td>
<td>94242</td>
<td>3.76124E-05</td>
</tr>
<tr>
<td></td>
<td>95241</td>
<td>8.23562E-05</td>
</tr>
<tr>
<td></td>
<td>95242</td>
<td>3.51934E-08</td>
</tr>
<tr>
<td></td>
<td>95243</td>
<td>2.73144E-06</td>
</tr>
<tr>
<td></td>
<td>96242</td>
<td>2.60530E-10</td>
</tr>
</tbody>
</table>
This is a partial list of all isotopes in material 51 which corresponds to rod number 101. The complete table 40 provides material compositions for all 264 rods.

<table>
<thead>
<tr>
<th>Cell</th>
<th>Gram density</th>
<th>Calculated volume</th>
<th>Mass</th>
</tr>
</thead>
<tbody>
<tr>
<td>101</td>
<td>1.04507E+01</td>
<td>1.93158E+02</td>
<td>2.01864E+03</td>
</tr>
<tr>
<td>105</td>
<td>1.04501E+01</td>
<td>1.93158E+02</td>
<td>2.01853E+03</td>
</tr>
<tr>
<td>109</td>
<td>1.04500E+01</td>
<td>1.93158E+02</td>
<td>2.01851E+03</td>
</tr>
<tr>
<td>113</td>
<td>1.04499E+01</td>
<td>1.93158E+02</td>
<td>2.01848E+03</td>
</tr>
<tr>
<td>117</td>
<td>1.04497E+01</td>
<td>1.93158E+02</td>
<td>2.01844E+03</td>
</tr>
</tbody>
</table>

Cell numbers 101, 105, 109, 113, and 117 correspond to rod numbers 101, 102, 103, 104, and 105 respectively. The complete table 50 provides this information for all 264 rods.

Figure 29. Workflow from NGSI spent fuel library to final transported spectra.

Table 2. Partial Table 50 from MCNP initialization from 4% initial enrichment, 15 GWd/MTU burnup and 5 year cooling time NGSI spent fuel assembly.
CHARACTERIZATION OF SPENT FUEL RODS AND ASSEMBLIES

This section describes applying the methodologies developed above to characterize spent fuel rods and assemblies. The neutron source components, spontaneous fission and (α,n), vary as a function of the fuel parameters, which include initial enrichment, burnup, and cooling time [36]. The gamma spectra has been studied through multivariate analysis to predict spent fuel parameters [37]. This chapter applies similar techniques to better understand the relationship between the neutron spectra and spent fuel parameters. These results are compared to previous investigations of neutron emissions from spent fuel assemblies.

Methods

The methodology developed in above formed the basis for characterizing spent fuel rods and assemblies. The NGSI Spent Fuel Library was used to obtain nuclide masses from individual rods within spent fuel assemblies of varying initial enrichments, cooling times, and burnups. ORIGEN-S used the nuclide mass data from these rods to calculate the neutron spectra for both spontaneous fission and (α,n) reactions. A MCNP geometry of a single spent fuel rod with cladding was created. Separate MCNP models were built for each rod using each of the following neutron source term options: total neutrons, spontaneous fission, and (α,n) reaction neutrons. MCNP transported one hundred million neutrons per source term with average surface flux (F2) tallies on the top plane, bottom plane, and cylindrical surface of each rod.

Computational simulations were run for a single rod from each of the assemblies in the NGSI spent fuel library, which included varying initial enrichments, burnups, and cooling times. The results and discussion below examine rods with an initial enrichment of 4 percent, burnups of 15, 30, and 45 GWd/MTU, and cooling times of 5, 20, 40, and 80 years. The available fuel parameters combinations in NGSI spent fuel library 2a are shown in Table 2.

Table 3. Available fuel parameter combinations from NGSI Spent Fuel Library 2a

<table>
<thead>
<tr>
<th>Initial Enrichment (wt%)</th>
<th>Burnups (GWd/MTU)</th>
<th>Cooling Times</th>
</tr>
</thead>
<tbody>
<tr>
<td>2%</td>
<td>15, 30</td>
<td>0d, 14d, 1y, 5y, 20y, 40y, 80y</td>
</tr>
<tr>
<td>3%</td>
<td>15, 30</td>
<td>0d, 14d, 1y, 5y, 20y, 40y, 80y</td>
</tr>
<tr>
<td>4%</td>
<td>15, 30, 45</td>
<td>0d, 14d, 1y, 5y, 20y, 40y, 80y</td>
</tr>
<tr>
<td>5%</td>
<td>15, 30, 45, 60</td>
<td>0d, 14d, 1y, 5y, 20y, 40y, 80y</td>
</tr>
</tbody>
</table>

Cases for 2 wt%, 45 and 60 GWd/MTU; 3 wt%, 45 and 60 GWd/MTU; 4 wt%, 60 GWd/MTU are not included as they are overly burned and not realistic.

A similar methodology was applied to complete 17x17 pressurized water reactor (PWR) assemblies. This was a significant increase in complexity as each assembly contained 264 rods. Figure 30 shows a complete 17x17 PWR assembly from the NGSI spent fuel library using the MCNPX Visual Editor. Unique source terms were generated for each rod. A single MCNP run contained complete geometry and materials definitions for the assembly but only one of the fuel rods as a source. A total of 264 MCNP runs per assembly were conducted and the tally totals on the outside of the assembly were added to simulate neutron emissions from an entire assembly.
Results and Discussion

This section begins with results and discussions from individual spent fuel rod characterization. The applicability of this technique to entire assemblies is examined. The results are compared to other spent fuel neutron emission calculations.

Individual Spent Fuel Rod Characterization

For all simulations of individual fuel rods, MCNP tallies passed all ten statistical checks. In general, the relative error was less than 1% for energy bins less than 8 MeV. Energy bins at higher energies had a larger relative error due to fewer neutrons at those energies. For example, relative error often exceeded 10% for energy bins above 13 MeV.

Figure 31 shows the total neutron spectra on the outside cylindrical surface of rods with an initial enrichment of 4 percent, burnup of 15 GWd/MTU and cooling times of 5, 20, 40, and 80 years. The total neutron flux decreases with increasing cooling time, however the peak around 3.0 MeV persists. Separating the total neutron flux into its components, as shown in Figure 32, shows the total neutron flux, spontaneous fission neutron flux and (α,n) reaction neutron flux emitted by the rods for each cooling time. The magnitude of the spectrum from spontaneous fission drops significantly with increasing cooling time, while the magnitude of the spectrum from (α,n) reactions is much less affected. Figure 33 shows the components of the total neutron flux for spent fuel rods with an initial enrichment of 4 percent, burnup of 30 GWd/MTU and cooling times of 5, 20, 40, and 80 years. In this case, the peak around 3.0 MeV starts to appear at the 40-year cooling time. However at 80 years the contributions from the (α,n) reactions is significant. Figure 33 shows the components of the total neutron flux for spent fuel rods with an initial enrichment of 4 percent, burnup of 45 GWd/MTU and cooling times of 5, 20, 40, and 80 years. At this higher burnup, the contribution from (α,n) reactions is minimal even at the 80-year cooling time.
Figure 31. Change in total neutron spectra for 4% initial enrichment and 15 GWD/MTU burnup spent fuel rods at various cooling times.

Figure 32. Change in neutron spectra by source for 4% initial enrichment and 15 GWD/MTU burnup spent fuel rods at various cooling times.
Integrating the energy spectra from each component of the neutron flux enables better visualization of the overall trend. Figure 34 shows the neutron fluxes separated into total neutron flux, spontaneous fission neutron flux and ($\alpha$,n) reaction neutron flux for the 4% initial enrichment and 15 GWd/MTU burnup spent fuel rods. As cooling time increases, the total neutron and spontaneous fission neutron fluxes both decrease, but the neutron flux from the ($\alpha$,n) reaction increases. In Figure 35 and Figure 36, which show 30 GWd/MTU and 45 GWd/MTU burnups, the total neutron and spontaneous fission neutron fluxes both decrease, while the ($\alpha$,n) reaction neutron flux only has a slight comparative increase. The difference in trends between the spontaneous fission and ($\alpha$,n) reaction neutron fluxes offers a way to predict the cooling time for known fuel burnup and initial enrichment.
Analysis of ORIGEN-S data shows that at lower cooling times most neutrons are produced from spontaneous fission of heavy nuclides with $^{244}$Cm as the largest contributor. However, the increase in ($\alpha$,n) reactions with increasing cooling times is due to the increase in $^{241}$Am which decays from $^{241}$Pu. As shown in Figure 37, $^{241}$Am is the major contributor to the ($\alpha$,n) reaction neutron source term in spent fuel. The increase in alpha emissions from $^{241}$Am results from an increase in ($\alpha$,n) reactions.

There is an important distinction between computational versus experimental measurements. Separating spontaneous fission neutrons and ($\alpha$,n) reaction neutrons in computational models is straightforward. A detector in the field would only be able to measure neutron counts and energies. Therefore, it was necessary to create a method to separate these two neutron sources from a detector measurement.
Figure 37. Change in neutron source strength as a function of cooling time for 4% initial enrichment and 45 GWd/MTU spent fuel rods.

Based on the characteristics of their spectra, the spontaneous fission neutrons and (α,n) reaction neutrons can be estimated from a total neutron spectrum. The spontaneous fission neutrons peak between 0.8 to 1 MeV and have a long tail extending to 12 to 15 MeV. Conversely, the spectrum for (α,n) reaction neutrons has a softer curve with a maximum between 2.5 and 3.0 MeV [16]. The (α,n) reaction neutron spectra tail ends at approximately 5 MeV.

Figure 38. Change in (α,n) reaction rate by α emitter as a function of cooling time for a 4% initial enrichment and 15 GWd/MTU burnup spent fuel rod.

Using these characteristics, the results are separated into two integrals. The first is from 0.1 to 1.6 MeV, covering most of the spontaneous fission neutrons. The second is from 1.6 MeV to 3.6 MeV, spanning most of the (α,n) reaction neutrons. Figure 38 shows the characteristic ranges on a graph of the total neutron spectrum for a spent fuel rod with 4 percent initial enrichment, 15 GWd/MTU burnup and a 40-year cooling time. These characteristic energy ranges consistently
captured about 50% of all spontaneous fission neutrons and 72% of all \((\alpha, n)\) reaction neutrons. The border at 1.6 MeV was chosen as this was the energy where the neutron flux from \((\alpha, n)\) reactions began to increase significantly. This technique requires a neutron detector capable of neutron spectroscopy at appropriate resolutions.

Figure 39 shows the energy regions of interest for spontaneous fission and \((\alpha, n)\) reactions plotted versus cooling time for spent fuel rods with 4% initial enrichment and 15 GWd/MTU burnup. The spontaneous fission flux decreases at a faster rate, which allows for characterizing the cooling time based on the ratio of the two fluxes. Each of these fluxes is dependent on exponential decay, with the spontaneous fission neutron flux decaying at a faster rate than the \((\alpha, n)\) reaction neutron flux. This allows characterizing the cooling times based on the ratio of the two fluxes. The functional form is expected to be exponential decay as shown in Figure 40.

Figure 41 shows the energy regions of interest for spontaneous fission and \((\alpha, n)\) reactions plotted versus cooling time for spent fuel rods with 4% initial enrichment and 30 GWd/MTU burnup. Figure 42 shows the energy regions of interest for spontaneous fission and \((\alpha, n)\) reactions plotted versus cooling time for spent fuel rods with 4% initial enrichment and 45 GWd/MTU burnup. For these two burnups, the characteristic energy ranges both decreased at similar rates, which did not allow characterizing the cooling time using this technique.

![Graph showing characteristic energy ranges for spontaneous fission and \((\alpha, n)\) neutrons.](image)

*Figure 39. Characteristic energy ranges for spontaneous fission and \((\alpha, n)\) neutrons shown for a 4\% initial enrichment, 15 GWd/MTU burnup and 40-year cooling time.*
Figure 40. Change in neutron flux in characteristic energy ranges as a function of cooling time for 4% initial enrichment and 15 GWd/MTU spent fuel rods.

Figure 41. Change in the (α,n) and spontaneous fission neutron flux ratio as a function of cooling time for 4% initial enrichment and 15 GWd/MTU spent fuel rods.
Applicability to Spent Fuel Assemblies

An examination of the neutron source spectra emitted from a complete assembly is necessary to determine if this method is still applicable. Figure 43 shows the comparison of a normalized neutron flux between a 4% initial enrichment, 15 GWd/MTU burnup individual fuel rod compared to a 17x17 PWR assembly with the same parameters. The overall qualitative agreement furthered applying the same technique to complete assemblies. The next step was to compare the neutron source terms for different cooling times. Figure 44 shows that the (α,n) reaction neutrons make up a larger percentage of the total neutron flux for an assembly with 4% initial enrichment, 15 GWd/MTU burnup, and 40-year cooling time compared to the assembly with 4% initial enrichment, 15 GWd/MTU burnup, and a 5-year cooling time. Based on these results, the method was found to be applicable to spent fuel assemblies. However, the challenge of using this method with spent fuel rods with higher burnups is expected to occur with spent fuel assemblies with higher burnups. This is due to relatively few (α,n) reaction neutrons compared to the total neutrons.

Figure 42. Change in neutron flux in characteristic energy ranges as a function of cooling time for 4% initial enrichment and 30 GWd/MTU spent fuel rods.

Figure 43. Change in neutron flux in characteristic energy ranges as a function of cooling time for 4% initial enrichment and 45 GWd/MTU spent fuel rods.
Comparison to Other Neutron Source Term Calculations

Table 4 shows the comparison between the GATORS methodology and the methodology in this work to calculate the spontaneous fission and \((\alpha,n)\) reaction neutron source term percentages for assemblies with varying spent fuel parameters.

Weldon et al. calculated that for an assembly with 4\% initial enrichment and 15 GWd/MTU burnup, the \((\alpha,n)\) emissions are slightly less than the spontaneous fission emissions at a 40-year cooling time and slightly more than the spontaneous fission emissions at an 80-year cooling time [48]. The same trend was identified in Figure 32 for transported spectra for an individual rod. While their work used NGSI SFL2c and this work used NGSI SFL2a, which had different shuffling schemes, the total neutrons per second emitted from assemblies of varying parameters was found to be similar.

Table 4. Comparison between spontaneous fission and \((\alpha,n)\) contributions to the neutron source term between Richard et al. and this work.

<table>
<thead>
<tr>
<th>Initial Enrichment</th>
<th>Burnup</th>
<th>Cooling Time</th>
<th>SF% and (\alpha) Harkness</th>
</tr>
</thead>
<tbody>
<tr>
<td>Richard et al.</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2%</td>
<td>15</td>
<td>5</td>
<td>95% and 5%</td>
</tr>
<tr>
<td>3%</td>
<td>30</td>
<td>5</td>
<td>98% and 2%</td>
</tr>
<tr>
<td>4%</td>
<td>45</td>
<td>5</td>
<td>99% and 1%</td>
</tr>
</tbody>
</table>

Summary

The linkage code developed for ORIGEN-S and MCNP6 allowed analysis of transported neutron source terms for spent fuel rods and assemblies from the NGSI spent fuel libraries. The identification of neutron energy spectral characteristics in individual rods and 17X17 PWR
assemblies enables predicting fuel parameters, such as cooling time, when initial enrichment and burnup are known. This method was most effective for spent fuel at lower burnups. The method faced difficulty at higher burnups with fewer \((\alpha,n)\) reaction neutrons compared to spontaneous fission neutrons. It was found that the transported neutron energy spectrum from an assembly had similar trends when compared to the non-transported neutron energy spectrum from other research.

Figure 45. Comparison of the neutron spectra for a 4% initial enrichment, 15 GWd/MTU burnup PWR assembly at 5-year cooling time (left) and a 40-year cooling time (right).
**Objective 3:**

*Construct a prototype instrument and use it to measure spent fuel casks at a commercial spent fuel storage facility.*

In the field of nuclear nonproliferation and safeguards, neutron multiplicity distributions and signatures of neutron and/or gamma-ray coincidence measurements are useful features for source identification and characterization. Liquid and plastic types of organic scintillators are widely used, since they are sensitive to both gamma rays and fast neutrons without using moderating materials. However, gamma-ray sensitivity can be a limitation in applications of high gamma-ray fields when measuring neutrons, in some of those situations, alternative detectors are needed.

The $^4$He detectors are known for their gamma-ray rejection capability and therefore, as detailed in this chapter, the feasibility of using them to measure neutron and gamma-ray time-dependent cross-correlation distributions is explored. The chapter begins with an introduction of the coincidence measurement and its applications, followed by a description of how to categorize various coincidence pairs, and concludes with an evaluation of the measured cross-correlation functions with different experimental setups. The shape of the cross-correlation functions can be an indicator of which type, and where the neutron source is. Thus, from this work, the application of the $^4$He detectors is extended to an area of nuclear safeguards where the identification of nuclear materials and material-geometry configuration assessment are highly desired.

**INTRODUCTION AND ADVANTAGES OF MEASURING CROSS-CORRELATION FUNCTIONS WITH $^4$HE DETECTORS**

Recent techniques based on coincidence measurement of neutrons and gamma rays, usually within a time window on the order of a few tens of nanoseconds, can be used for identifying and characterizing nuclear materials. The cross-correlation functions represent signatures allowing identification of typical neutron sources (i.e. spontaneous-fission or ($\alpha$, n)) [38], radioactive material-geometry configuration [39], and special nuclear material (uranium, plutonium) quantification [40]. Most researchers have been focused on organic liquid or plastic scintillators. However, during cross-correlation measurements, high-Z materials have to be employed to shield overwhelming gamma rays and achieve better ratio of different cross-correlation functions. That results in additional uncertainties that are introduced from scattered and induced neutrons.

To obtain comparable numbers of correlated neutrons and gamma rays without extended measurement times or gamma-ray shielding, the novel $^4$He fast neutron scintillation detectors are used to fulfil the above-mentioned requirements. In this work, the charge-integration based pulse shape discrimination technique is utilized to discriminate between neutrons and gamma rays to identify the four possible correlated pairs: ($\gamma$, $\gamma$), ($n$, $\gamma$), ($\gamma$, $n$), and ($n$, $n$), where the first index corresponds to detector #1 and the second corresponds to detector #2 in any given cross correlation function. Two neutron sources, a $^{252}$Cf spontaneous-fission and a Pu-Be ($\alpha$, n) neutron source, are used to measure the cross-correlation functions at various source-detector distances. Both the total and individual cross-correlation functions are obtained, and their shapes can be easily distinguished among different neutron sources. The ($n$, $n$) and ($\gamma$, $n$) correlations are selectively analyzed and the peak position of the ($n$, $n$) and ($\gamma$, $n$) pairs shows a linear correlation with the source-detector distance. In particular, the ($n$, $n$) correlated data can be very clearly assayed even in presence of significant gamma-ray fields without the use of shielding.
By analyzing the signatures in the cross-correlation distributions, the $^4$He detectors can be used as a tool to characterize potentially unknown nuclear materials with simplified setups and reduced uncertainties.

**CROSS-CORRELATION FUNCTIONS WITH $^{252}$CF SPONTANEOUS NEUTRON SOURCE**

The experimental setup of the cross-correlation functions measurement involves two identical $^4$He detectors and two neutron sources, a 73.7 $\mu$Ci $^{252}$Cf spontaneous-fission source and a 1 Ci Pu-Be ($\alpha$, n) source. In this section, we will start with the spontaneous-fission source.

The detectors are placed in parallel with a detector-to-detector distance of 30 cm. Five source locations are selected. The source is initially placed between the detectors at three source-detector distances. Both symmetric and asymmetric configurations are investigated as follows: “15cm-15cm”, “10cm-20cm”, and “5cm-25cm”. Additionally, two more source locations are chosen along the y-axis (see the experimental setup in Figure 46. The source is placed at the centroid of the two detectors to explore the effect of detectors locus-dependent response on the measured cross-correlation functions. During all the measurements, no gamma-ray shielding is used, yet reasonable neutron coincidence counts are achieved without additional uncertainties being introduced.

![Figure 46. Experimental setup of the cross-correlation functions measurements.](image)

Each $^4$He detector has two output signals from the photomultiplier tubes (PMTs). Therefore four PMT anode signals are fed into the Struck 14-bit 250MHz multi-channel waveform digitizer in total. One thousand data points are recorded for each pulse, giving a total pulse acquisition time window of 4 $\mu$s. The digitizer’s constant fraction discriminator is enabled, and a trigger is generated when the actual trapezoidal value goes below the half of its maximum value (default setting). Thus a better pulse timing resolution (~2 ns) could be obtained by using the linear interpolation method, which is a better resolution than the digitizer inherent 4 ns sampling interval. The average timestamp of the two PMT's outputs from one detector is calculated and will be used to determine the time differences between coincidence events between the two detectors.

The above-mentioned four categories of correlated pairs: ($\gamma$, $\gamma$), (n, $\gamma$), ($\gamma$, n), and (n, n) are identified through the charge-integration based PSD. As discussed in Chapter 3, neutrons tend to have larger delayed scintillation light yield than gamma rays, and the final PSD plot is shown in Figure 47. For low-energy neutrons, the output pulses usually exhibit low amplitude, therefore those cannot be discriminated as readily from the gamma-ray events. However, they only account
for 11% of the total number of pulses. Events above the PSD line are regarded as neutrons while events that fall into the lower region are regarded as gamma rays. The same aforementioned pulse filtering algorithms [89] were applied to remove pile-up events and other anomalous events to improve the neutron-gamma-ray identification accuracy. Cross-correlation functions are calculated by determining the time differences between the two detectors within a coincidence time window (80 ns utilized here) at a specific pulse amplitude threshold (30 mV), corresponding to incoming neutrons of approximately above 0.5 MeV to be detectable.

For the 73.7 μCi $^{252}\text{Cf}$ spontaneous-fission source, during a 3.2-hours measurement, 9326 correlated events are recorded by the two detectors. If using four detectors instead of two, the same statistics would be achieved in one-sixth the time due to higher correlation possibilities. Figure 48 shows the measured cross-correlation functions at various source-detector distances (“15cm-15cm” on the left and “10cm-20cm” on the right). Neutrons are emitted with a characteristic fission energy distribution resulting in a wider neutron time-of-flight (TOF) spread, thus the $(\gamma, \gamma)$ peak has a smaller time spread than the $(n, n)$ peak. In addition, significantly less $(\gamma, \gamma)$ pairs are counted than the $(n, n)$ pairs, which validates the high gamma-ray rejection ability of the $^4\text{He}$ detectors. The two side peaks are produced when a fission neutron arrives in coincidence with a gamma ray, which is categorized as $(n, \gamma)$ or $(\gamma, n)$ pairs.

Figure 47. Scatter plot of the integration of fast versus slow component of the cross-correlation measurement.

Figure 48. Measured cross-correlation functions at various source-detector distances.
Figure 48. Measured $^{252}$Cf cross-correlation functions at “15cm-15cm” source-detector distance (left) and “10cm-20cm” source-detector distance (right). The total cross-correlation function is obtained by summing all the correlated pairs together. Uncertainty is shown on the “total” curve, and is of identical magnitude in the individual component curves for data points at the same vertical position (“Normalized counts”-amplitude) as the total curve.

Ideally, the $(\gamma, \gamma)$ peak should occur at time zero, however due to variance in the scintillation photon transport time in the detectors and electronic delays in the PMTs and cables, a 1 ns shift of the $(\gamma, \gamma)$ distribution is observed. The time shift is corrected during later analysis. Comparing the two figures in Figure 48, when the detectors have the same distance to the source, the cross-correlation distributions are symmetric around time zero. When the source is moved between the detectors (i.e. along the x-axis indicated in Figure 47), notable changes in terms of peak position and time spread are observed due to the differences in neutron travel path length. These changes provide the basis to characterize material-geometry configurations.

Noted in previous cross-correlation measurements with organic scintillators [39], some spurious peaks are observed in $(n, \gamma)$ and $(\gamma, n)$ cross-correlation functions. These spurious peaks are due to misclassified events, for example, an additional neutron interacting with one of the detectors when a $(\gamma, \gamma)$ pair was already being counted. The pulse from the above mentioned double-event has gamma-ray timing features but is more likely to be classified as a neutron due its large scintillation light output. Increasing detector distance or applying high-Z gamma-ray shielding materials could be one possible way to reduce the accidentals, by sacrificing detection efficiency and accuracy. In contrast, the $^4$He detector produced relatively clean $(n, \gamma)$ and $(\gamma, n)$ correlation curves void of such misclassification features justifying its advantages for this application.

Gamma rays are near instantaneous with low detection efficiency and are not significantly affected by the source location, and it is also hard to characterize the peak position of the coincident mixed neutrons and gamma rays due to their reduced statistics, and their energy-dependent combined detection efficiency. Thus, the $(n, n)$ cross-correlation function is the best source to characterize the source location. Figure 49 shows the measured and Gaussian-fitted time delay distributions for $^{252}$Cf $(n, n)$ pairs at various source-detector distances (left), and the relationship between the $(n, n)$ peak position and source location (right).

Figure 49. Measured and Gaussian-fitted time delay distributions for $^{252}$Cf $(n, n)$ pairs at various source-detector distances (left), and the $(n, n)$ peak position as a function of source location (right) after gamma peak correction.
In addition to changing the source locations, five thresholds in analog-digital-converter (ADC) unit are tested during post pulse processing as 0 V (no increased threshold), 0.05 V, 0.1 V, 0.15 V, and 0.2 V. As shown in Figure 50, increasing the threshold would reject more low-amplitude-events, and these events are also harder to be identified through PSD. As a result, more “true neutron events” are preserved and the time delay distribution of the (n, n) pairs tends towards a Gaussian distribution, indicated by the decreasing full width at half maximum (FWHM). However, as the threshold keeps going up, there will be less events of sufficient amplitude to be kept and the filter becomes too conservative to ensure good statistics. Therefore, it is necessary to choose a suitable threshold based on the required goodness of fit and the coincidence count rate of each measurement. For this work, 0.05 V threshold is selected and used in all the analysis.

![Figure 50. Measured and Gaussian-fitted time delay distributions for $^{252}$Cf (n, n) pairs at various ADC thresholds at “15cm-15cm” source-detector distance (left), and the FWHM of Gaussian fitting as a function of digitizer threshold at “15cm-15cm” source-detector distance (right).](image)

When moving the $^{252}$Cf along y-axis, theoretically there should be no shift in the measured cross-correlation functions. However, as illustrated previously, there are differences in the PMT timing characteristics, which are associated with photon transport, source locations, and the dual-PMT read-out. Therefore, slight shifts are observed depending on where in the detector the interaction took place. As shown in Error! Reference source not found., the (n, n) peak position shows a linear trend when the source is moved along the detector’s active volume. The results indicate this type of cross-correlation measurement may potentially provide source depth information which allows a comprehensive geometry characterization when leveraging multiple detector locations of volumetric distributed sources such as small mixed waste drums. The y-axis dependence could also be derived from detection statistics if moving the detectors along the sample-location in the y-axis in a controlled manner. Yet as observed from Figure 51, the linear relationship is relative weak and the change in the (n, n) peak position is relatively small. Therefore, if measurement condition allows, we recommend rotating the source container to replicate the same behavior as in Figure 49. Otherwise, simply rotating the detectors would not allow a full mapping of the source container.
CROSS-CORRELATION FUNCTIONS WITH PU-BE (Α, N) SOURCE

The experiment with the Pu-Be (α, n) source is similar as the 252Cf measurement, and the 1 Ci Pu-Be source is placed at the same source locations. For reference purpose, source characterization is performed at first with an EJ-309 liquid scintillator. For this specific Pu-Be source, within 3 seconds, a total of 50526 even events are recorded at an 80 keVee threshold. Among those, 50% events are neutrons and 50% events are gamma rays after PSD analysis. The same neutron/gamma-ray ratio is obtained at all suitable thresholds, indicating a relative strong and consistent spectrum response between the two types of radiation. The Pu-Be source emits only one neutron per (α, n) reaction, while 252Cf has an average neutron multiplicity of 3.76 from spontaneous-fission events [41]. Thus, the fraction of measured neutrons-correlations will be lower than for 252Cf, as one can tell from the measured cross-correlation functions visually.

Figure 52 shows the measured cross-correlation functions at “15cm-15cm” source-detector distance, where 21206 correlated events are measured with in 0.87 hour. The total cross-correlation function from the 252Cf measurement is also plotted here for comparison. In contrast to the 252Cf cross-correlation functions, less (n, n) and (γ, γ) pairs are recorded in the Pu-Be measurement, while (n, γ) and (γ, n) pairs mainly contribute to the shape of the total cross-correlation function. In addition to observing the peaks from plots, Table 5 below shows the ratio of different pairs at “15cm-15cm” source-detector distance for 252Cf and Pu-Be. The differences in the contribution of each type of pairs can be used when identifying nuclear materials [42], and future work will be focused on developing advanced source identification/classification methods.
Figure 52. Measured Pu-Be and $^{252}$Cf cross-correlation functions at “15cm-15cm” source-detector distance. Data from Pu-Be and $^{252}$Cf is normalized by “per ns” Uncertainty is showed on the “total” Pu-Be curve, and is of identical magnitude in the individual curves for data points at the same vertical position (“Normalized counts”-amplitude) as the total curve.

Table 5. The ratio of the four categories pairs from $^{252}$Cf and Pu-Be measurements.

<table>
<thead>
<tr>
<th>Pairs (%)</th>
<th>$^{252}$Cf</th>
<th>Pu-Be</th>
</tr>
</thead>
<tbody>
<tr>
<td>(n, n)</td>
<td>50.87 ± 0.11</td>
<td>34.64 ± 0.94</td>
</tr>
<tr>
<td>($\gamma$, $\gamma$)</td>
<td>9.92 ± 0.40</td>
<td>10.82 ± 0.48</td>
</tr>
<tr>
<td>($\gamma$, n)</td>
<td>19.63 ± 0.59</td>
<td>26.02 ± 0.80</td>
</tr>
<tr>
<td>(n, $\gamma$)</td>
<td>19.58 ± 0.59</td>
<td>28.52 ± 0.84</td>
</tr>
</tbody>
</table>

Similarly to the $^{252}$Cf measurements, the Pu-Be measurement shows ($\gamma$, $\gamma$) pairs are still near instantaneous and of insignificant magnitude, and the (n, n) pairs have a relative even distribution since only one neutron is emitted per ($\alpha$, n) reaction. On the other hand, ($\gamma$, n) pairs (or (n, $\gamma$) pairs) have enough counts with relatively small spurious peaks, therefore they can be used when characterizing the source location. Similar to the $^{252}$Cf measurement, when moving the source between the two detectors, it still shows a linear correlation (within uncertainty) between the ($\gamma$, n) peak position and source location. The lack of clear features in the (n, n) cross-correlation means that the ($\gamma$, n) data was a better option for source location determination in case of the Pu-Be source. There is a small (n, n) cross-correlation bump near $\Delta t=0$, originating from induced fissions in the plutonium. Certain discrepancies could be found when trying to fit the ($\gamma$, n) peak position and source location linearly, while such discrepancies are not observed in the $^{252}$Cf measurement. For the ($\gamma$, n) pairs. When moving the source, the differences in $\Delta t$ are relatively small, since only one neutron’s travel time varies with the change in distance. In addition, gamma rays have higher yield and energy for the Pu-Be source than $^{252}$Cf [43] [44], and are more likely to be misclassified as neutrons due to pile-up and other factors. Therefore, slight nonlinearity is observed as on the right of Figure 53.
Figure 53. Measured and Gaussian fitted time delay distributions for Pu-Be (γ, n) pairs at various source-detector distances (left), and the (γ, n) peak position as a function of source location (right) after gamma peak correction.

**4He CROSS-CORRELATION MEASUREMENT SYSTEM CONCLUSIONS**

To conclude the results and discussions above, for the first time, the time-dependent cross-correlation distributions from a $^{252}$Cf spontaneous-fission source and a Pu-Be ($\alpha$, n) sources are measured using the $^4$He fast neutron detectors. Pulses are recorded using a Struck digitizer with trigger timing algorithm enabled to achieve an enhanced timing resolution. An offline charge integration based PSD technique is performed to discriminate between neutrons and gamma rays. Both separate ((γ, γ), (n, γ), (γ, n), and (n, n)) and total cross-correlation functions are measured at various source-detector distances.

At equal source-detector distance, the cross-correlation distributions are symmetric around time zero. When moving the source between the two $^4$He detectors, notable detectable changes are observed. The peak position of Gaussian-fitted time delay distributions ((n, n) for $^{252}$Cf and (γ, n) for Pu-Be) shows a clear linear correlation with the source location, which provides the basis for locating nuclear materials in larger geometrical samples. The same trends are observed when the sources are moved along the detector’s active chambers, indicating the 3D characterization potentials. In addition, the shapes of the cross-correlation functions for $^{252}$Cf and Pu-Be shows distinct features due to different contributions from the correlated pairs. Source identification can be achieved by leveraging that difference.

Comparing with previous cross-correlation measurements, no shielding materials are needed due to the $^4$He detector’s acceptable fast neutron detection efficiency and lower gamma-ray sensitivity than organic materials. Therefore, introducing $^4$He detectors to cross-correlation measurements can result in reduced uncertainties, fast measurement times, and expanding the feasibility in high gamma-ray fields. The results demonstrate the feasibility of using $^4$He detectors to measure total and individual cross-correlation functions from both spontaneous-fission and ($\alpha$, n) neutron sources. The detector exhibits its unique characteristics that can be leveraged as advantages over the widely-used organic scintillators in identifying and characterizing nuclear materials using the cross-correlation functions. Future work includes measuring actual radioactive waste barrels, the development of source identification and location-mapping methods, as well as other applications in nuclear nonproliferation and homeland security.
Objective 4:

Develop the capability of cask imaging through a combination of neutron and gamma measurements.

CHARACTERIZATION OF FAST NEUTRON EMISSIONS FROM SPENT FUEL IN DRY CASK STORAGE

This section describes the characterization of the fast neutron emissions from spent fuel assemblies in dry cask storage. The structure of the HI-STORM 100S dry cask system is discussed. The original methodology was expanded to incorporate up to 32 assemblies and the complexity of the cask structure. The final simulations used a more detailed geometry and neutron source than was found in past research, which homogenized portions of the fuel assemblies and cask. The previously identified technique using the differences in energies between spontaneous fission and ($\alpha$,n) reaction neutrons was applied to the neutron energy spectra emitted from the cask. Full cask and partial cask loadings were simulated to determine if it is possible to identify diverted assemblies using count rate data. Results were compared to prior research on fingerprinting techniques by others.

HI-STORM 100S Dry Cask System

Holtec International currently serves 53 nuclear facilities in the United States (over half of the entire installed base) and 24 international facilities [45]. Due to the widespread use, the Holtec HI-STORM 100S system was selected for this research. It accommodates a wide variety of spent nuclear fuel assemblies by utilizing different multipurpose canisters with identical exterior dimensions inserted into a common overpack. Detailed drawings and information regarding this system are available in the Final Safety Analysis Report for the HI-STORM 100 Cask System [46].

All multipurpose canisters (MPCs) have identical external dimensions. The structural components of the MPC are fabricated from various steels including types 316, 316LN, 304, and 304LN. The fuel basket itself is made of steel. Metamic, a boron carbide metal matrix composite material, is placed on two sides of each assembly to absorb neutrons. The inside of the canister is filled with helium because it is inert, has excellent heat transfer capabilities, and allows the use of helium leak-detection methods to ensure proper sealing [47].

Each MPC consists of a fuel basket, base plate, canister shell, lid, and a closure ring. The rectilinear grid design of the MPC fuel baskets offer the benefit of uniform distribution of the metal mass and efficient radiation attenuation [46]. The MPCs have different internal layouts to accommodate different numbers and types of fuel assemblies. For example, the MPC-24 contains a maximum of 24 PWR assemblies, the MPC-32 contains a maximum of 32 PWR fuel assemblies, and the MPC-68 contains a maximum of 68 BWR assemblies. This research focuses on the MPC-32 due to the economic savings of higher density storage for PWR assemblies.

The HI-STORM 100S overpack consists of inner and outer cylindrical steel shells filled with concrete. The primary function of the carbon steel is to provide structural stability and the primary function of the concrete is to provide shielding against gamma and neutron radiation [46]. Both steel and concrete have a long history of usage in nuclear applications. It has a heavy bolted concrete and steel lid. The cask provides adequate shielding to absorb radiation and structural strength necessary to protect against natural disasters or accident scenarios.
Methods

The computational methodology discussed previously was expanded to incorporate up to 32 assemblies inside a cask system. This detailed modeling was automated through development of a MATLAB program. For each of the 32 assembly locations in the MPC-32, the program allowed choosing to insert an assembly from the NGSI spent fuel library or to leave that location empty to mimic a diverted assembly. The program then referenced ORIGEN-S output files and built a complete MCNP geometry of assemblies with appropriate neutron sources inside of a cask. The final simulation had more detailed geometry and neutron sources than past research methods which homogenized portions of the fuel assemblies and cask.

The MCNP models of the HI-STORM 100S cask system are based on the geometry developed in the neutron fingerprinting research by Rauch [48]. The MCNP model included the MPC, baseplate, pedestal, platform, inner shell, concrete shield, outer shell, lid top plate, lid shield block and radial plates. Assembly details included the guide/instrument tubes, gaps between fuel and cladding, Metamic, and basket steel. These assembly details provide a realistic basis for the transport and shielding calculations in MCNP. Neutron detectors were modeled to obtain realistic detection system statistics. The models were based on the helium-4 detectors and had stainless-steel cylinders with an active length of 20 cm and an inner diameter of 4.4 cm for a total active volume of 304 cm³. Figure 54 shows two cross sections of the MCNP geometry. Figure 55 shows a single Westinghouse 17x17 PWR assembly inside the MPC.

![Figure 54. Horizontal and vertical cross sections of the MCNP model of the HI-STORM 100S cask system. Helium-4 detectors are spaced equally around the cask.](image)
Simulations were performed with up to 32 assemblies inside of a HI-STORM 100S dry cask system. All ORIGEN-S calculations were performed on a standard desktop computer running the Windows operating system. All MCNP simulations were performed on the University of Florida HiPerGator computing cluster. ORIGEN-S calculations were almost instantaneous. MCNP calculations took about two days for 10 billion initial neutrons. Chen et al. note that the limiting factors in a complicated shielding problem are how realistic the calculation model can be \cite{49}. Reliable source term estimation and high-fidelity geometry modeling are critical. ORIGEN-S was used to calculate a reliable source term estimate. The geometry included appropriate details for accurate shielding and transport calculations.

Initial cask loadings used assemblies with identical parameters from the NGSI spent fuel library to create the easiest scenario for detecting a diverted assembly. Additional loadings were developed based on real loading data from ORNL. Average surface flux tallies (F2) were used on the inside and outside cylindrical surfaces of the canister and overpack. Average cell flux tallies (F4) were used for the gas volume of each of the helium-4 gas scintillation detectors.

Unlike many simulations which homogenize the fuel, the fuel assembly structures are modeled explicitly in this work. The NGSI spent fuel libraries have each rod as a single axial fuel region so it was not possible to calculate axial source term variation. For actual PWR assemblies, the axial burnup is relatively flat in the axial midsection with under-burned fuel for the top and bottom 30 cm of the assembly \cite{50}. Although the neutron energy spectra were calculated individually for each rod in the 17x17 PWR assemblies, there were only minor differences in the spectra due to different localized burnups. To simplify MCNP input files, the spectra for all 264 rods was averaged and set as the energy distribution for all rods in that assembly. This approach introduced a minor approximation for the radiation transport as normally each rod would have a slightly different energy distribution.

Initial sensitivity analyses started with identical assemblies inside the HI-STORM 100 dry cask system. These had the same materials, neutron source strength, and neutron spectra emitted. The symmetry allows picking five different assembly diversion locations (1, 2, 6, 7, and 13) that are
equivalent to removing any of the 32 assemblies. The MPC-32 assembly layout with these representative locations are shown in Figure 56. Additional simulations were run with assemblies of varying average neutron source strengths to approximate real cask loadings. These were used to determine the effect on the neutron flux measured by the detectors in the same azimuthal area.

![Figure 56. The loading pattern of 32 spent fuel assemblies in a HI-STORM 100S cask.](image)

Results and Discussions

The first results discussed are from applying the previously identified spent fuel characterization technique to neutron energy spectra data emitted from the cask. The second results discussed are from techniques to identify diverted assemblies using count rates.

Spent Fuel Characterization Through Spectral Data

As my previous research showed, the \((\alpha, n)\) reaction neutrons have a maximum between 2.5 and 3.0 MeV. My prior technique estimated the number of spontaneous fission neutrons and \((\alpha, n)\) reaction neutrons based on the characteristics of their spectra: spontaneous fission neutrons peaked between 0.8 to 1 MeV and \((\alpha, n)\) reaction neutrons peaked between 2.5 and 3 MeV. This section discusses the feasibility of applying this method to spent fuel assemblies in dry cask storage.

The tally on the MPC shows the neutron spectra with self-shielding from the assemblies but without the attenuation from the concrete overpack. Figure 57 shows the neutron flux as it crosses the inside cylindrical surface of the MPC. The spectrum inside the cask is affected by self-shielding from the assemblies as it is qualitatively different than the flux from individual rods. Figure 58 shows the neutron flux on the outside cylindrical surface of the cask. The large peak at approximately 2.3 to 2.4 MeV was not observed in individual rod fluxes which led to investigating the isotopic composition of the concrete overpack. Concrete is approximately 50 weight percent oxygen. The total neutron cross section for \(^{16}\text{O}\) has a valley at similar energies and that results in a peak in the emitted neutron spectra. The difference between the internal and external neutron fluxes is approximately 5 orders of magnitude, illustrating the strong radiological shielding from the concrete overpack.
In general, the relative error was less than 10% for energy bins less than 5 MeV. As expected, the relative error increased for higher energy bins with fewer particles. Relative error often exceeded 20% for energy bins above 8 MeV.

![Graph](image1.png)

*Figure 57. Surface flux tally (F2) on inside cylindrical surface of MPC.*

![Graph](image2.png)

*Figure 58. Surface flux tally (F2) on outside cylindrical surface of concrete overpack.*

In all cases, over 80% of the neutrons escaping the overpack were below 200 keV. These bins are not shown in figures to enhance visibility of the spectral characteristics at fast energies. As seen in Figure 59, the total flux was strongly linked to the burnup and cooling time of the spent fuel. Otherwise, the overall shapes of each neutron spectra otherwise greatly resemble each other. In comparison, the neutron spectra from individual rods and assemblies had various peaks and valleys depending on the spent fuel parameters.
This behavior was theorized to be due to neutron attenuation from the cask system and self-shielding from the surrounding assemblies. In the context of nuclear engineering, attenuation is measured by cross sections. The total neutron cross-section ($\sigma_T$) is the likelihood that a neutron of a certain energy will interact with a target nucleus. These cross-sections can vary greatly with the kinetic energy of the neutron. The effective removal cross section ($\Sigma_R$) is the probability that a fast neutron undergoes a first collision which removes it from the group of uncollided neutrons [51]. The effective removal cross section ($\Sigma_R$) of concrete is almost constant for fast neutrons from 2 to 12 MeV [52]. Previous research has calculated the fast neutron effective removal cross section for various concretes and found that hydrogen and oxygen are major contributors [53]. Self-shielding from the spent fuel assemblies attenuates and scatters the neutrons resulting in spectra with magnitude as the only distinguishing characteristic.

![Figure 59](image1.png)

*Figure 59. Neutron spectra emitted from casks containing spent nuclear fuel assemblies of varying parameters.*

![Figure 60](image2.png)

*Figure 60. Change in neutron emissions on the outside cylindrical surface of the cask containing spent fuel assemblies with parameters of 4% initial enrichment and 15 GWd/MTU burnup as a function of cooling time.*
The same technique developed in previous sections was applied to the neutron emissions from a cask. The spent fuel assemblies inside had an initial enrichment of 4 percent, burnup of 15 GWd/MTU and cooling times of 5, 20, 40, and 80 years. Figure 60 shows the tally on the outside cylindrical surface of the overpack separated into total neutron flux, spontaneous fission neutron flux and (α,n) reaction neutron flux for different cooling times. As cooling time increases, the total neutron and spontaneous fission neutron fluxes both decrease, but the neutron flux from the (α,n) reaction increases. This is the same trend that was seen in individual spent fuel rods in Chapter 4.

The fluxes for the initial characteristic energy ranges (0 to 1.6 MeV and 1.6 to 3.6 MeV) are shown in Figure 61 and Figure 62 for 15 GWd/MTU and 45 GWd/MTU fuel. Figure 63 shows the alpha to SF ratio versus cooling time using the initial characteristic energy ranges. The initial characteristic energy ranges do not show an identifiable trend and would not be a reliable method to determine the cooling time of spent fuel. In contrast, Figure 41 shows an identifiable trend for the alpha to SF ratio versus cooling time for an individual fuel rod. The lack of an identifiable trend for spent fuel inside a cask was attributed to the high degree of moderation from the HI-STORM 100S cask system. However, higher energy neutrons may pass through the dry cask without scattering that could provide more useful characteristic energy range data. To test this theory, the characteristic range for spontaneous fission neutrons was adjusted higher in energy range from 3.6 MeV to 5.0 MeV. Figure 64 and Figure 65 show the same 15 GWd/MTU and 45 GWd/MTU burnups with the new energy ranges. The alpha to SF ratio shows a weak correlation to the cooling time as shown in Figure 66. Although an improvement from the initial energy ranges, the weak correlation would be difficult to use for characterization purposes due to the precise statistics needed.

![Figure 61. Neutrons emissions using initial characteristic energy ranges for spent fuel assemblies with parameters of 4% initial enrichment and 15 GWd/MTU burnup.](image-url)
Figure 62. Neutrons emissions using initial characteristic energy ranges for spent fuel assemblies with parameters of 4% initial enrichment and 45 GWd/MTU burnup.

Figure 63. Alpha/SF ratio using initial characteristic energy ranges as a function of cooling time for assemblies of various burnups.
Figure 64. Neutrons emissions using revised characteristic energy ranges for spent fuel assemblies with parameters of 4% initial enrichment and 15 GWd/MTU burnup.

Figure 65. Neutrons emissions using revised characteristic energy ranges for spent fuel assemblies with parameters of 4% initial enrichment and 45 GWd/MTU burnup.
Additionally, the technique faces challenges determining the cooling time for fuel at higher burnups due to the limited \((\alpha, n)\) reaction neutron contribution to the total neutron source. Average burnup was around 35 GWd/MTU two decades ago and is over 45 GWd/MTU today. Examination of the MCNP output files shows that spontaneous fission emissions dominate the total neutron signal at burnups greater than 30 GWd/MTU and cooling times less than 80 years. These results agree with conclusion from the study on total neutron emissions for the NGSI spent fuel libraries by Weldon et al which found \(^{244}\text{Cm}\) dominated the spectrum [36].

Based on these results, using the prior technique to determine the cooling time of spent fuel assemblies inside a cask is not feasible due to the HI-STORM 100S overpack resulting in qualitatively similar energy spectra. Additionally, utilities using fuel to higher burnups reduces the number of \((\alpha, n)\) reaction neutrons for measurement worsening the signal to noise ratio for the comparison of the \((\alpha, n)\)-to-spontaneous fission neutrons. Thus, efforts were shifted towards methods utilizing total neutron fluxes which do not rely on energy discrimination.

Identification of Diverted Assemblies Using Count Rates

The sensitivity analysis results were completed for each of the diversion scenarios. Neutron fluxes were normalized per source neutron. Diverting an assembly results in less self-shielding of neighboring assemblies. Each of the 100 detector locations measures neutron fluxes around equal azimuthal angles around the cask. Figure 67 shows that outer fuel assemblies obscure the diversion of an inner fuel assembly. There is a substantial change in symmetry for the Detectors 71 through 80 are located close to the missing assembly locations. Figure 68 shows the flux change compared to a fully loaded cask for these detectors when an assembly is missing from one of the characteristic locations. These detectors measure approximately 6\% lower flux when an assembly at position 1 is missing and approximately 8\% lower flux when an assembly at position 2 is missing compared to a fully loaded cask. However, when an assembly at positions 6 or 7 is missing, the change in flux is less than 1\%. Detecting a diverted assembly at positions 6 or 7 is unlikely. When an assembly at position 13 is missing, the detectors measure approximately 2\% higher than flux than a fully loaded cask. In addition, this is as close to an ideal scenario as possible with identical
assemblies, neutron source strengths, and spectra. A real cask would have varying source strengths that would lead to more difficult analyses. Based on these results, it is possible to detect a diverted assembly at positions 1 or 2 due to the flux decrease. It would be difficult to detect a diverted assembly at the other locations.

The work by Rauch found that a diverted assembly at positions 1 and 6 could be detected, position 2 might be able to be detected, and positions 7 and 13 cannot be detected by analyzing signatures through his acceptance criteria [28]. This previous work supports the conclusion that detection of assemblies that have been diverted around the outside perimeter is easier than detection of diverted inner assemblies.

Figure 67. Fast neutron fluence at each detector from full cask and casks with assemblies removed from 5 different locations (left). Data subset zoomed into the area showing the fluence differences (right).

Figure 68. Percentage change in flux for each missing assembly location. Error bars are standard error from MCNP.

The next set of simulations compared real cask loading data from ORNL to the identical assembly loadings. The loading pattern and average neutron source terms are shown in Figure 69. These three loading patterns provided a chance to test the proposed diversion detection method with real source term values. However, the ORNL loading patterns did not have isotopic data, so
the NGSI spent fuel libraries were still used as these should have similar isotopic information. The energy spectrum chosen was that from a 4% initial enrichment, 45 GWD/MTU burnup and 5-year cooling time as there was no energy spectrum provided for the ORNL data.

Figure 69. Loading pattern and average neutron source terms (n/s) per assembly for three real casks provided by ORNL [48].

The fluence measured by each detector is sensitive to the neutron source strength of individual assemblies as shown in Figure 70. Although the general shape of the signatures is similar, each loading pattern has a peak in a different quadrant. While Figure 67 showed variations in symmetry for each quadrant that could identify a diverted outer assembly, the ORNL loadings were fully loaded and there was a lack of symmetry between quadrants. Therefore, symmetry alone cannot be relied upon to identify diversions.
Identifying diversions through comparisons of expected to actual flux values at specific detector locations or sets of locations is a feasible technique. However, there is a lack of expected flux values for most, if not all, casks today. Measurements are not taken upon loading and this chapter has described the time-consuming process to generate MCNP simulations of a cask. The next section discusses a potential solution that mathematically calculates the expected flux values at specific detector locations using information about the fuel assemblies and cask.

A simulation of 10 billion initial neutrons represents about 2 seconds of real activity for a cask containing 32 assemblies with an average burnup of 45 GWd/MTU and a 5-year cooling time. Although there are 10 billion initial neutrons, due to further prompt fissions, delayed fissions, and \((n,xn)\) reactions, MCNP tracks approximately 14 billion neutrons. Only about 7 million neutrons actually escape the sides of the cask. It was not necessary to simulate longer than two seconds as statistical results were acceptable. Standard errors were less than 10% and the tally fluctuation chart bins passed all 10 built-in statistical checks.

An appropriate detector would need to have enough neutrons exiting the cask to measure with reasonable statistics. The total surface area of the outside of the cask walls is approximately \(5.76 \times 10^5\) cm\(^2\). Ten detectors, each with a 50 cm\(^2\) surface area, would cover 0.09% of the cask. For the highest neutron flux case of a 45 GWd/MTU burnup with 5 years of cooling time, these detectors would be experiencing 3000 n/s across the whole energy range. For the lowest neutron flux case of a 15 GWd/MTU burnup with 80 years of cooling time, these detectors would be experiencing 15 n/s across the whole energy range. These simulations serve as a proof of concept for neutron spectroscopy of spent fuel inside a dry cask using an appropriate detector.

**Summary**

This study investigated the characterization of neutron emissions from a HI-STORM 100S dry cask storage system with spent fuel assemblies of varying parameters inside. Although a significant neutron flux exits the dry cask storage system, neutrons at lower energies dominate the spectrum. While higher energy neutrons are emitted, the lower source strength presents a challenge for obtaining reasonable statistics. One set of simulations applied energy discrimination techniques.
to determine the cooling time of spent fuel inside a cask. The other set of simulations investigated using neutron count rates to identify diverted assemblies.

The first simulations examined if it was possible to determine the cooling time of assemblies inside the cask based on the energy of neutrons emitted from the cask. This utilized the previous spontaneous fission versus ($\alpha$,n) reaction neutron technique. However, all neutron energy spectra measured on the outside of the cask appear qualitatively similar due to self-shielding from the assemblies and attenuation from the concrete overpack. Other possible energy ranges were tested. Although they produced a weak correlation, the reliance on higher energy neutrons would need longer measurement times to obtain reasonable statistics. This method was determined to not be feasible for spent fuel in a dry storage cask.

The second simulations investigated the possibility of identifying diverted assemblies using count rates. By comparing the relative neutron fluxes from a fully loaded cask to a partially loaded cask, the simulations showed the ability to identify diverted assemblies on the outside perimeter. However, diversion of interior assemblies is more difficult to identify due to minimal changes in the neutron flux.

The results were compared to previous research regarding the neutron emissions from spent fuel. Differences were noted in which specific assembly locations could be detected if diverted, however the same conclusion was reached: diverted assemblies are easier to locate when they are on the outside perimeter.

**PREDICTING EXTERNAL NEUTRON COUNT RATES USING OPERATOR DECLARED INFORMATION**

This work analyzes a series of typical spent fuel loadings with the goal of determining the individual contribution of each spent fuel assembly to the neutron flux measured outside the cask. A method for predicting the flux at specific locations outside the cask using parameters specific to the cask and assembly loading patterns is also discussed. This provides a solution to a significant shortcoming of before-and-after fingerprinting techniques, the requirement of having a before fingerprint for successful comparison.

**Methods**

There were 32 separate simulations with a single assembly contributing to the neutron source term. This allowed quantitatively determining the relative contribution from each assembly to the total neutron flux outside the cask that would be measured by each detector. All 32 assemblies were from the NGSI Spent Fuel Library and had 4% initial enrichment, 45 GWd/MTU burnup and a 5-year cooling time. Each of the 32 simulations was run for 1 billion neutrons with a surface tally around the outside cylindrical surface of the cask and cell flux tallies for each of the 100 detectors.

Assemblies will be referred to by the numbering shown in Figure 68. The neutrons emissions from three real cask loadings provided by ORNL were also simulated. The ORNL data included a loading pattern and average neutron source terms (n/s) per assembly that was then incorporated into the HI-STORM 100S dry cask system geometry built in MCNP. The ORNL data also included initial enrichment and maximum burnup values for each assembly. The closest matching assembly from the NGSI spent fuel library was inserted into the simulations at that assembly location. For example, an ORNL assembly with 42.69 GWd/MTU burnup and an initial enrichment of 3.65% had a matching NGSI assembly of 45 GWd/MTu and an initial enrichment of 4%.
Results and Discussions

The difficulty of identifying diverted inner assemblies is due to the self-shielding from the outer assemblies. Quantifying the contribution of each assembly to the overall neutron source term is the first step towards determining the sensitivity required to identify diversions. Figure 72 shows the contribution to the overall neutron source term from each assembly normalized to unity. These results are from an average surface flux tally (F2) in MCNP. There is a strong symmetry in the expected contribution for each assembly. Each assembly around the perimeter contribute slightly less than 5% of the total neutrons with about 2% of the neutrons coming from each assembly in the next layer inward. The innermost assemblies contribute only about 1% each to the total neutrons.

Figure 72. Contribution from each assembly to the total neutron flux on the outside cylindrical surface of the cask.
A helium-4 detector on the outside of the cask would be receiving neutrons from all the assemblies inside, however some assemblies would contribute more neutrons than others. Figure 73 shows the contribution from each assembly to a detector located immediately to the right of the cask. Approximately 67% of the neutrons measured by the detector come from the four outermost assemblies (10, 16, 22 and 28). Approximately 24% of the detector come from the 6 assemblies in the next column (4, 9, 15, 21, 27 and 32). The remaining 9% of the neutrons come from the remaining 22 assemblies.

Figure 74. Contribution from each assembly to a single detector located at the upper right of the cask.
The neutron contributions from each assembly to a detector at the upper right of the cask is shown in Figure 74. Approximately 50% of the neutrons measured by the detector come from the two closest assemblies (28 and 32). The next greatest contributor is assembly 27, which contributes 13% of the neutrons.

The last analysis compared the predicted flux to the flux calculated by MCNP for three cask loadings provided by ORNL as shown in Figure 75. The neutron flux can be predicted with three parameters: (1) geometric effect from the dry cask system and self-shielding from the assemblies; (2) relative contributions from each assembly for the specific azimuthal detector location; and (3) loading pattern with average neutron source strength per assembly. The attenuation effect from the dry cask system is the flux measured by each detector using the cask loaded with identical assemblies. The attenuation effect has a periodic behavior as shown in Figure 76. Flux measurements from the detectors follow a periodic function as you move around the cask due to self-shielding from the assemblies and cask attenuation effects.

![Figure 75. Loading pattern and average neutron source terms (n/s) per assembly for three real casks provided by ORNL [48].](image)
Figure 76. Attenuation from the cask system and assemblies at specific detector locations.

Figure 77 shows the matrix multiplication needed to calculate the external neutron flux around the cask at 100 specific detector locations. Figure 78 shows the actual (MCNP) flux versus predicted (mathematical) measurements at detectors around a single quadrant of the cask. The predicted fluxes overestimated the flux from MCNP for most detector locations, however the relative magnitudes and overall trends are correct. Since the values are normalized, the diagonal locations were underestimated. The greatest change in the flux from one detector to the next occurs at the diagonal locations. Practical application of this technique would require crucial placement of detectors to ensure they are not placed between two measurement points.

Figure 77. Matrix multiplication needed to calculate the external neutron flux at 100 detector locations around the cask.
This work provides one possible solution to the lack of a prior fingerprint existing for casks. While these results are specific to the HI-STORM 100S cask system, the techniques are applicable to any spent fuel storage cask. MCNP models exist for other dry cask systems, such as the TN-32 cask [55]. If additional sensitivity is desired, another dimension could be added to the parameters, such as neutron energy or variations in flux along the z-axis of the cask. Additional dimensions may require more measurements and would involve more data analysis.

Figure 78. Actual (MCNP) versus predicted (mathematical) neutron flux for detectors 64 to 87.

Summary
This study created a way to mathematically model the fingerprint of a spent fuel cask using parameters specific to the cask and operator declared information about the fuel. Computational methods were developed to calculate these parameters using the HI-STORM 100S dry cask system and the Next Generation Safeguards Initiative Spent Fuel Libraries. The neutron flux can be predicted with three parameters: (1) geometric effect from the dry cask system and self-shielding from the assemblies; (2) relative contributions from each assembly for the specific azimuthal detector location; and (3) loading pattern with average neutron source strength per assembly.

Real assembly loading patterns from Oak Ridge National Lab were used to compare the quick mathematical model to full MCNP simulations. It was found that the predicted fluxes had the same relative magnitude and overall trends as full MCNP simulations. Practical application of this technique would require precise detector placement to ensure it is not between two measurement points as even a few degrees off can result in a significantly different neutron flux.

This work provides one possible solution to the lack of a prior fingerprint existing for casks. In addition, this technique is readily applicable to any spent fuel storage cask. Further refinements to this technique could include adding neutron energy or z-axis variations as an additional dimension to the parameters.
PUBLICATIONS AND PRODUCTS

Journal publications (7)


Conference proceedings (15+)


**Presentations & posters (15+)**


- Y. Liang, X. Wen, J. E. Baciak, A. Enqvist, Measurement of Neutron and Gamma-ray Cross-correlation Functions with 4He Detectors, SORMA-2018, Ann Arbor, MI, 11-14 June 2018


**Degrees/Theses (3)**

**PhD:**

- Characterization and Application Of 4He Fast Neutron Scintillation Detectors to Nuclear Materials and Radiation Detection, Yinong Liang

- Safeguards Approaches for Spent Nuclear Fuel in Dry Cask Storage, Ira A. Harkness III

**MSc:**

- Neutron Spectroscopy And Characterization Of Non-Extended And Extended Helium-4 Silicon-Photomultipliers, Cathleen Barker
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